Supporting Information

Non-monotonic Composition Dependence of the Breakdown of Stokes−Einstein Relation for Water in Aqueous Solutions of Ethanol and 1-Propanol: Explanation using Translational Jump-diffusion Approach

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S1. Supporting Tables and Figures

Table S1. The summary of the simulated systems and the production run for analyses for different compositions and temperatures.

	Simulation time (ns)					
T (K)	x_E =0.0	x_E =0.1	x_E =0.2	x_E =0.5	x_E =0.8	x_E =1.0
225	200	200	200	200	200	200
235	200	200	200	200	200	200
245	200	200	200	200	200	200
255	200	200	200	200	200	200
265	120	120	120	120	120	120
275	120	120	120	120	120	120
285	120	120	120	120	120	120
300	120	120	120	120	120	120

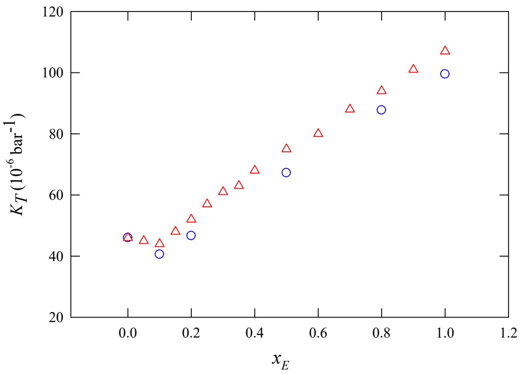

Figure S1. Composition dependence isothermal compressibility of W/E mixtures at 300K. The red (triangle) shows the isothermal compressibility of literature data[1] and whereas the corresponding blue (circle) simulated data of water-ethanol mixture at different mole fractions.

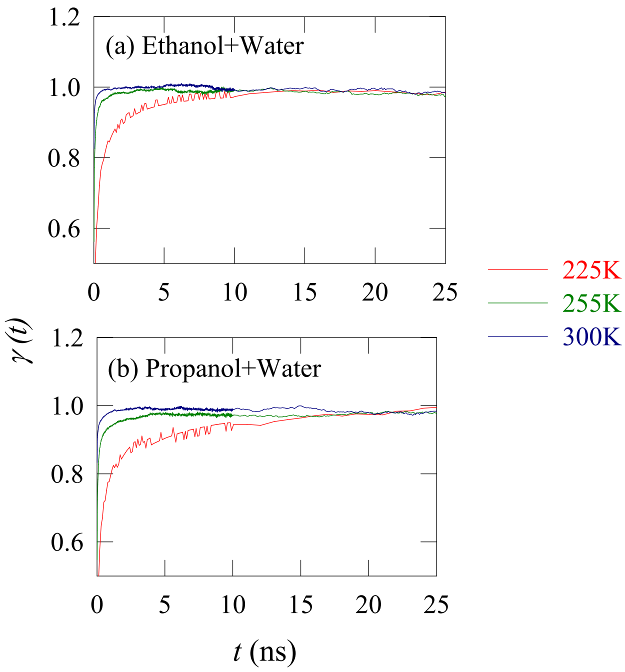

Figure S2. The exponent $\gamma(t)$calculated using the equation $\gamma(t) = d\ln\langle r^2(t)\rangle / d\,ln(t)$ for (a) E/W and (b) P/W binary mixtures for alcohol mole fraction x=0.2 at three different temperatures 225K, 225K, and 300K.

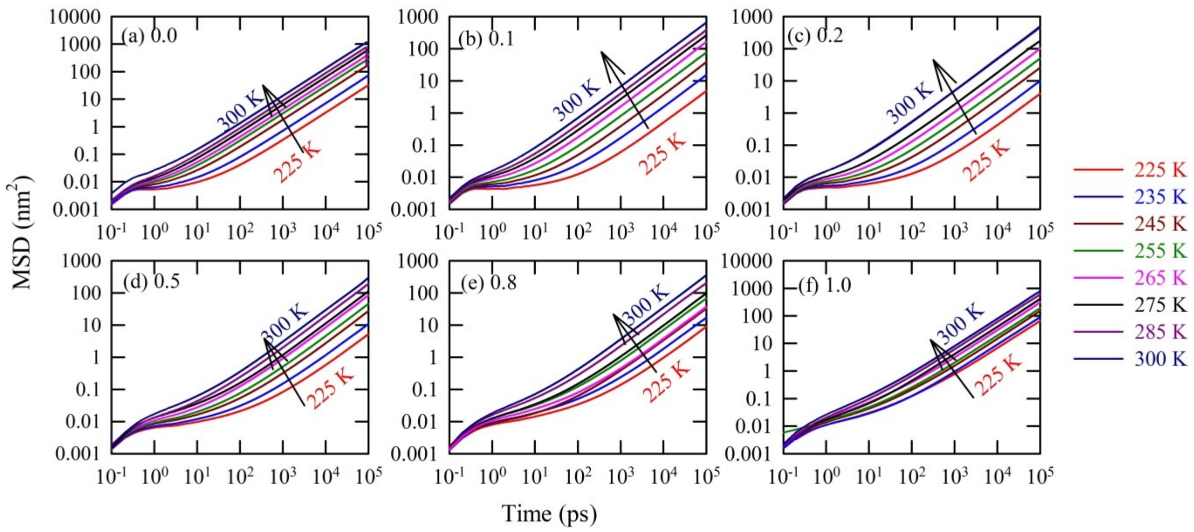

Figure S3. MSDs for the overall system at different temperatures and compositions of the W/E binary mixture.

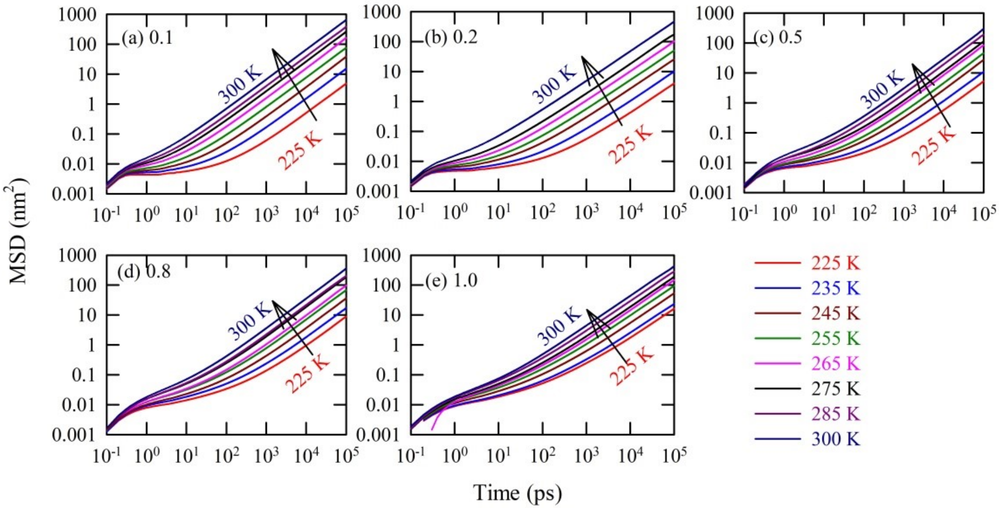

Figure S4. MSDs for the overall system at different temperatures and compositions of the W/P binary mixture.

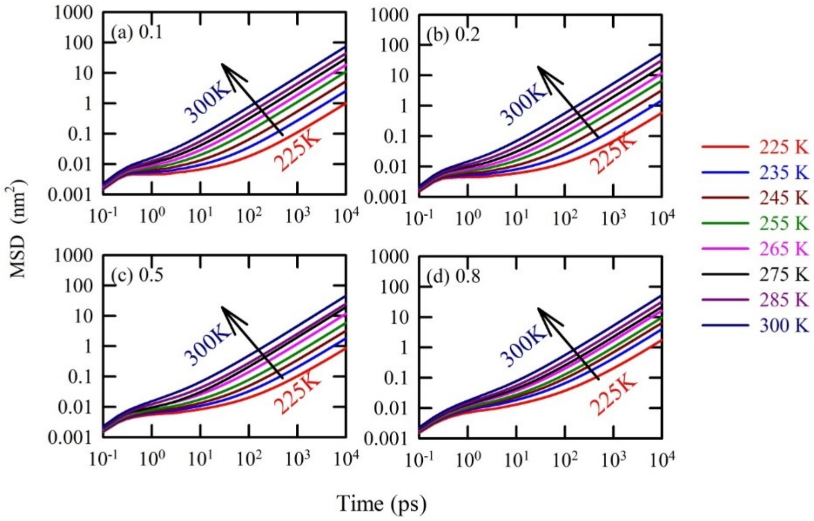

Figure S5. MSDs of water at different temperatures and compositions of W/E binary mixture.

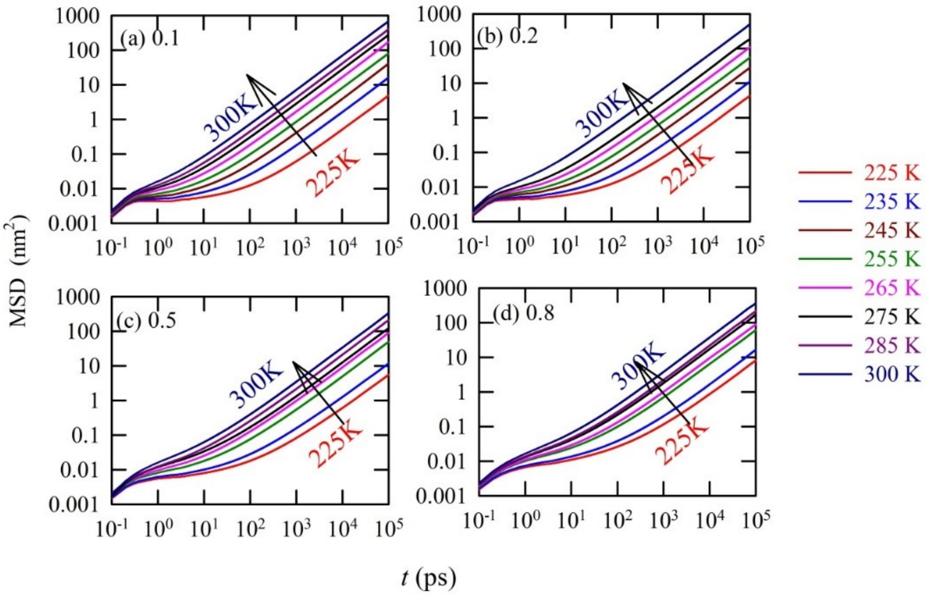

Figure S6. MSDs of water at different temperatures and compositions of W/P binary mixture.

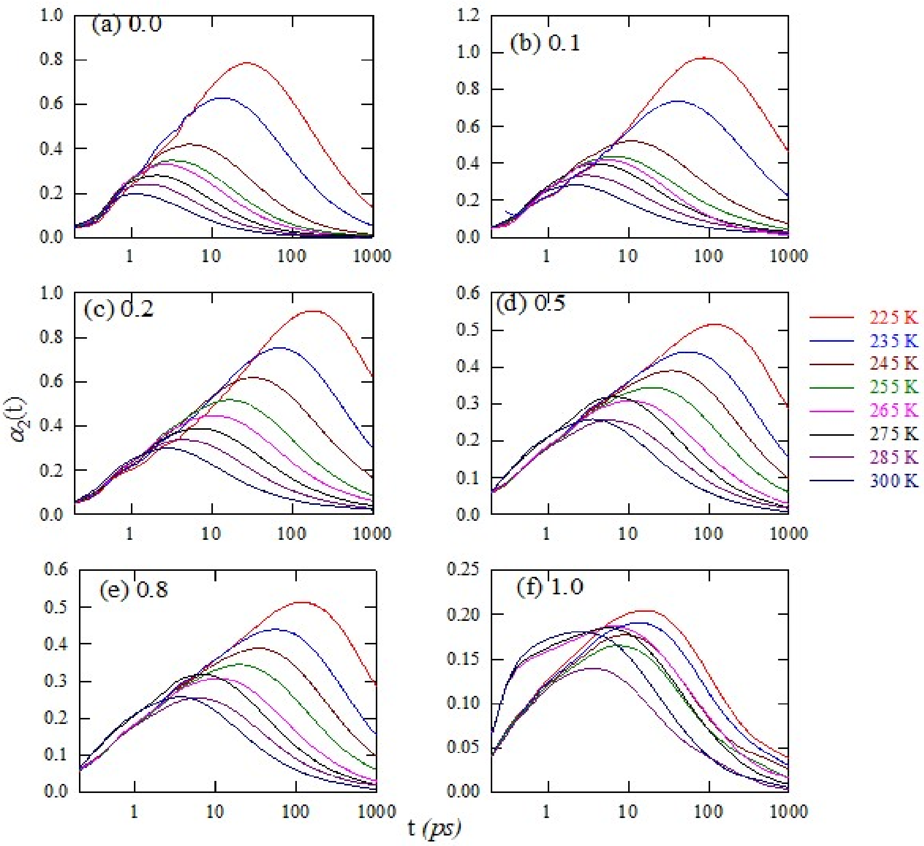

Figure S7. The average non-Gaussian parameter $\alpha_2(t)$ for the overall system at different temperatures and compositions of W/E binary mixture.

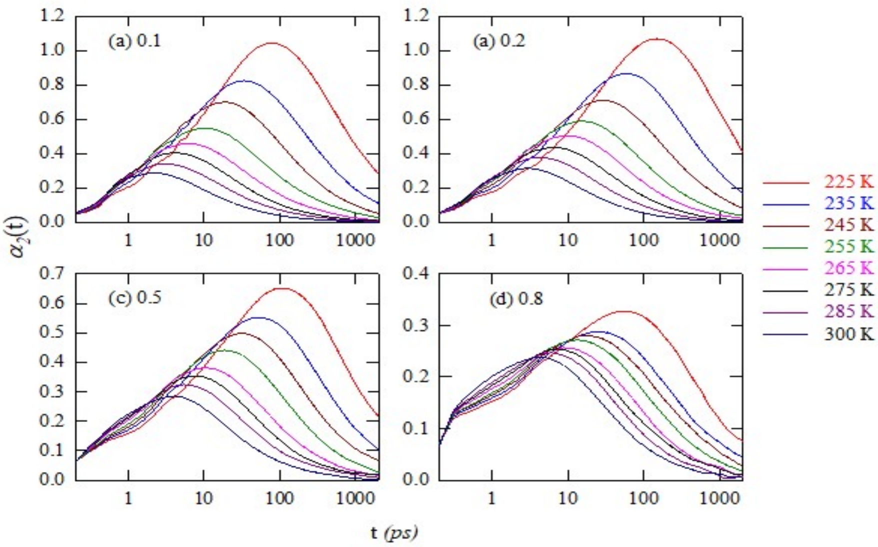

Figure S8. The non-Gaussian parameter $\alpha_2(t)$ for water at different temperatures and compositions of W/E binary mixture.

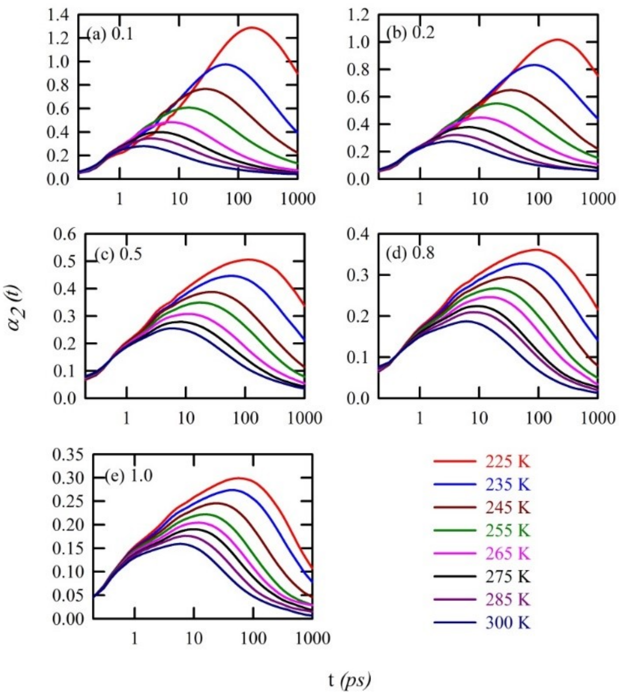

Figure S9. The average non-Gaussian parameter $\alpha_2(t)$ for the overall system at different temperatures and compositions for W/P binary mixture.

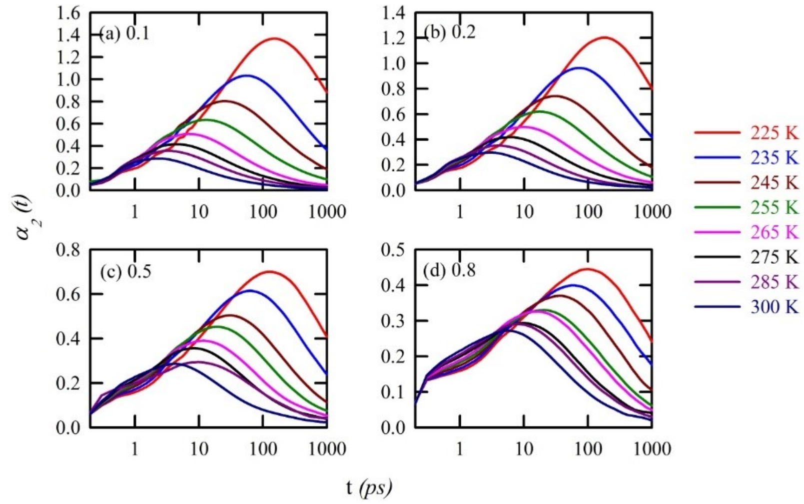

Figure S10. The non-Gaussian parameter $\alpha_2(t)$ for water at different temperatures and compositions of the W/P binary mixtures.

Table S2. The results of overall W/E system at different binary mixture of compositions diffusion coefficient D, t^*, r_2, jump-diffusion coefficient D_{Jump}, number of jumps n_{Jump}, the percentage contribution of jump χ_{Jump}, jump frequency v_{Jump}, percentage of jump trajectory segment P_{jump} and viscosity. The values in parenthesis are the standard errors of D and η.

Average (W/E)											
x_E = 0.0											
T(K)	**D /10^{-5} (cm^2/sec)**	**t^***	**r_2**	**D_{Jump} /10^{-5} (cm^2/sec)**	**D_{Res} /10^{-5} (cm^2/sec)**	**n_{Jump}**	**λ^2_{Jump}(Å^2)**	**contribution of jump χ_{Jump}**	**Jump Frequency v_{Jump} (ns^{-1})**	**Percentage of Jump trajectory Segment $P_{Jump} = \frac{n_{Ju}}{n_w t_{tr}}$**	**Viscosity η (cp)**
225	0.053(1.00×10^{-3})	27.25	0.22	0.016	0.037	207326	6.89	30.19	1.38	3.77	46.39(0.87)
235	0.128(1.80×10^{-3})	13.05	0.22	0.028	0.1	371875	6.84	21.88	2.48	3.22	15.83(0.3)
245	0.323(3.28×10^{-3})	5.39	0.23	0.041	0.282	535487	6.88	12.69	3.57	1.89	5.88(0.08)
255	0.540(4.60×10^{-3})	3.24	0.23	0.058	0.482	760191	6.80	10.74	5.07	1.62	3.37(0.08)
265	0.729(1.16×10^{-2})	2.62	0.24	0.075	0.654	620567	7.28	10.29	6.21	1.61	2.49(0.045)
275	1.120(1.81×10^{-2})	2.12	0.26	0.062	1.058	431909	8.59	5.54	4.32	0.91	1.66(0.02)
285	1.583(1.19×10^{-2})	1.60	0.27	0.056	1.527	371472	9.00	3.54	3.71	0.59	1.40(0.01)
300	2.469(3.84×10^{-2})	1.10	0.30	0.019	2.45	213386	10.60	0.77	1.07	0.12	0.793(0.11)
x_E = 0.1											
225	0.018(4.00×10^{-4})	85.70	0.22	0.006	0.0117	68421	7.47	33.90	0.46	3.93	138.43(10.78)
235	0.043(1.47×10^{-3})	41.07	0.24	0.011	0.0318	118283	7.99	25.70	0.79	3.25	54.77(1.22)
245	0.093(4.50×10^{-4})	20.11	0.24	0.022	0.071	242103	7.97	23.66	1.61	3.25	23.54(0.71)
255	0.194(2.77×10^{-3})	10.95	0.25	0.033	0.1612	241513	8.17	16.99	2.42	2.63	11.36(0.33)
265	0.336(4.56×10^{-3})	6.29	0.25	0.046	0.2901	334797	8.20	13.69	3.35	2.08	6.38(0.11)
275	0.535(3.50×10^{-3})	3.87	0.24	0.073	0.4624	568576	7.76	13.63	5.69	2.16	3.98(0.09)
285	0.800(9.81×10^{-3})	3.05	0.26	0.077	0.7228	542394	8.50	9.63	5.42	1.63	2.72(0.03)
300	1.378(1.30×10^{-2})	2.25	0.28	0.086	1.2917	532427	9.72	6.24	5.32	1.17	1.61(0.04)
x_E = 0.2											

225	0.009(1.60×10^{-4})	181.10	0.24	0.003	0.00634	33291	8.38	32.11991	0.22	4.04	253.60(17.31)
235	0.027(3.00×10^{-4})	68.64	0.24	0.007	0.0203	76412	8.25	25.64103	0.51	3.51	77.95(1.89)
245	0.054(7.80×10^{-4})	33.29	0.24	0.014	0.0395	150493	8.17	26.16822	1.00	3.33	36.56(2.069)
255	0.112(9.49×10^{-4})	16.96	0.25	0.024	0.0881	168144	8.44	21.40946	1.68	2.85	17.09(0.5)
265	0.196(1.46×10^{-3})	10.95	0.26	0.032	0.1641	217842	8.88	16.3182	2.18	2.38	9.37(0.086)
275	0.317(6.05×10^{-3})	6.67	0.26	0.050	0.2674	351098	8.58	15.75299	3.51	2.32	5.94(0.14)
285	0.535(4.69×10^{-3})	4.30	0.26	0.069	0.4663	465276	8.87	12.88997	4.65	2.00	3.51(0.04)
300	0.907(1.07×10^{-2})	2.85	0.28	0.074	0.8332	442111	10.09	8.156966	4.42	1.24	1.95(0.04)
						x_E = 0.5					
225	0.015(3.62×10^{-4})	118.30	0.27	0.005	0.0096	42060	9.82	34.24658	0.28	3.34	86.90(5.88)
235	0.035(8.19×10^{-4})	54.80	0.27	0.009	0.0256	87812	9.68	25.714	0.59	3.22	36.14(2.62)
245	0.059(5.75×10^{-4})	34.50	0.27	0.013	0.0459	120968	9.99	22.07131	0.81	2.79	21.22(0.69)
255	0.103(1.77×10^{-3})	19.81	0.28	0.020	0.0825	121184	10.12	19.5122	1.21	2.40	12.84(0.3)
265	0.226(2.79×10^{-3})	10.44	0.28	0.039	0.1865	225562	10.35	17.2949	2.26	2.35	5.85(0.19)
275	0.309(7.67×10^{-3})	7.16	0.28	0.049	0.2597	280571	10.44	15.87302	2.81	1.99	4.54(0.08)
285	0.448(5.34×10^{-3})	5.72	0.29	0.062	0.3862	344154	10.75	13.83311	3.44	1.96	3.18(0.05)
300	0.862(8.38×10^{-3})	3.72	0.30	0.091	0.7713	461247	11.84	10.55317	4.61	1.71	1.78(0.03)
						x_E = 0.8					
225	0.038(7.57×10^{-4})	61.61	0.33	0.007	0.0311	46280	14.03	18.3727	0.31	1.91	25.74(0.43)
235	0.094(1.77×10^{-3})	29.26	0.35	0.013	0.081	79539	15.18	13.82979	0.53	1.55	9.98(0.57)
245	0.141(2.60×10^{-3})	19.31	0.32	0.029	0.1118	189850	13.49	20.59659	1.27	2.44	6.84(0.2)
255	0.233(3.91×10^{-3})	14.00	0.33	0.035	0.1983	157033	13.53	15.00214	1.57	2.20	4.19(0.058)
265	0.364 (6.24×10^{-3})	9.50	0.36	0.035	0.3285	130972	15.91	9.628611	1.31	1.23	2.82(0.06)
275	0.485(6.19×10^{-3})	7.32	0.35	0.042	0.4428	165971	15.09	8.663366	1.66	1.21	2.30(0.05)
285	0.694(1.36×10^{-2})	6.00	0.36	0.050	0.6437	189561	15.94	7.207727	1.90	1.12	1.61(0.02)
300	1.096(1.41×10^{-2})	4.20	0.37	0.067	1.0292	236518	17.03	6.112023	2.37	0.99	1.10(0.02)
						x_E = 1.0					
225	0.133(1.63×10^{-3})	16.00	0.33	0.021	0.1122	140576	13.20	15.76577	0.94	1.50	5.67(0.09)
235	0.167(2.09×10^{-3})	13.25	0.33	0.019	0.1481	128110	13.62	11.37044	0.85	1.13	5.53(0.34)
245	0.295(2.57×10^{-3})	8.97	0.35	0.023	0.2715	141089	14.57	7.809847	0.94	0.84	2.98(0.05)
255	0.363(5.20×10^{-3})	7.17	0.33	0.034	0.3293	226810	13.49	9.358657	1.51	1.07	2.56(0.054)
265	0.564(8.18×10^{-3})	6.37	0.36	0.037	0.5271	138744	16.11	6.559121	1.39	0.87	1.68(0.02)
275	0.796(1.07×10^{-2})	6.91	0.40	0.043	0.7526	130187	19.61	5.404726	1.30	0.90	1.27(0.05)
285	1.105(1.48×10^{-2})	3.73	0.37	0.060	1.0454	216226	16.51	5.427899	2.16	0.80	1.00(0.01)
300	1.507(2.01×10^{-2})	4.10	0.41	0.061	1.4456	174283	21.06	4.048852	1.74	0.72	0.77(0.02)

Table S3. The results of water at different binary mixture compositions diffusion coefficient D, t^*, r_2, jump-diffusion coefficient D_{Jump}, number of jump n_{Jump}, the percentage contribution of jump χ_{Jump}, jump frequency ν_{Jump}, and percentage of jump trajectory segment P_{jump}. The values in parenthesis are the standard error of D.

				Water (W/E)						
T (K)	***D* /10^{-5} (cm^2/sec)**	***t** ps**	***r$_2$* (nm)**	***D$_{Jump}$* /10^{-5} (cm^2/sec)**	***D$_{Res}$* /10^{-5} (cm^2/sec)**	***n$_{Jump}$***	**λ^2_{Jump} (Å^2)**	**contribution of jump χ_{Jump}**	**Jump Frequency ν$_{Jump}$ (ns^{-1})**	**Percentage of Jump trajectory Segment $P_{Jump} = \frac{n_{Jump}}{n_w t_{traj}/t^*} \times 1$**
				x_E = 0.1						
225	0.023(4.25×10^{-4})	77.30	0.22	0.006	0.017	71608	7.16	33.90	0.53	4.11
235	0.055(3.25×10^{-3})	33.71	0.22	0.014	0.041	153417	7.29	32.71	1.14	3.84
245	0.106(6.85×10^{-4})	18.30	0.23	0.023	0.083	246540	7.66	25.16	1.83	3.35
255	0.215(2.83×10^{-3})	10.23	0.24	0.037	0.178	383681	7.83	19.46	2.84	2.90
265	0.368(5.90×10^{-3})	6.30	0.25	0.049	0.319	323865	8.16	14.64	3.60	2.27
275	0.570(7.13×10^{-3})	3.96	0.25	0.062	0.508	403015	8.33	11.58	4.48	1.75
285	0.855(2.55×10^{-2})	3.10	0.26	0.081	0.774	507740	8.60	10.00	5.64	1.75
300	1.432(2.87×10^{-2})	2.22	0.28	0.093	1.339	521073	9.64	6.75	5.79	1.27
				x_E = 0.2						
225	0.011(2.31×10^{-4})	150.6	0.23	0.004	0.007	33405	7.84	42.83	0.28	4.22

235	0.030(1.58×10^{-3})	58.30	0.23	0.008	0.022	76143	7.74	28.57	0.64	3.71
245	0.060(2.20×10^{-3})	29.28	0.23	0.016	0.044	142503	7.82	28.27	1.19	3.47
255	0.122(1.00×10^{-3})	15.36	0.24	0.026	0.096	155626	8.13	21.87	1.95	2.98
265	0.221(1.49×10^{-3})	9.96	0.25	0.036	0.185	199719	8.63	17.05	2.50	2.47
275	0.346(3.75×10^{-2})	6.21	0.25	0.056	0.29	318715	8.43	16.64	3.98	2.47
285	0.576(8.53×10^{-3})	4.10	0.26	0.075	0.501	413221	8.75	13.15	5.17	2.12
300	0.997(9.78×10^{-3})	2.88	0.27	0.094	0.903	466636	9.62	9.51	5.83	1.63
x_E = 0.5										
225	0.015(3.25×10^{-4})	107.00	0.25	0.004	0.011	20919	9.05	26.49	0.28	3.00
235	0.034(2.63×10^{-3})	52.36	0.25	0.008	0.026	42037	9.00	22.60	0.56	2.83
245	0.058(1.05×10^{-3})	33.18	0.26	0.013	0.045	58940	9.62	21.70	0.79	2.59
255	0.109(1.19×10^{-3})	18.89	0.27	0.020	0.089	90820	9.78	17.99	1.21	2.28
265	0.220(1.07×10^{-2})	10.44	0.28	0.036	0.184	106543	10.09	15.96	2.13	2.22
275	0.308(1.42×10^{-2})	8.06	0.29	0.043	0.265	118160	10.92	13.93	2.36	1.89
285	0.459(1.12×10^{-2})	5.46	0.28	0.061	0.398	174733	10.52	13.15	3.49	1.89
300	0.888(2.71×10^{-2})	3.910	0.31	0.085	0.803	210185	12.19	9.50	4.20	1.64
x_E = 0.8										
225	0.029(6.45×10^{-4})	57.46	0.29	0.005	0.024	8361	11.09	15.43	0.28	1.60
235	0.071(6.77×10^{-3})	25.13	0.30	0.0097	0.0619	15174	11.62	12.05	0.51	1.27
245	0.111(9.77×10^{-3})	33.00	0.26	0.021	0.09	34528	10.96	17.00	1.15	2.03
255	0.175(1.70×10^{-3})	13.71	0.30	0.028	0.147	28370	11.67	12.00	1.42	1.95
265	0.287(1.85×10^{-2})	9.17	0.33	0.029	0.258	24977	13.97	7.98	1.25	1.14
275	0.411(1.48×10^{-2})	6.74	0.32	0.041	0.37	37449	13.23	8.46	1.87	1.26
285	0.552(2.65×10^{-2})	5.92	0.34	0.055	0.497	44754	14.62	9.36	2.24	1.32
300	0.968(7.13×10^{-2})	4.45	0.36	0.060	0.908	42283	16.97	5.94	2.11	0.93

Table S4. The results of overall water/1-propanol system at different binary mixture compositions diffusion coefficient D, t^*, r_2, jump-diffusion coefficient D_{Jump}, number of jump n_{Jump}, the percentage contribution of jump χ_{Jump}, jump frequency ν_{Jump}, and percentage of jump trajectory segment P_{jump} and viscosity. The values in parenthesis are the standard errors of D and η.

Average (W/P)											
T(K)	**D /10^{-5} (cm²/sec)**	**t^* (ps)**	**r_2 (nm)**	**D_{Jump} /10^{-5} (cm²/sec)**	**D_{Res} /10^{-5} (cm²/sec)**	**n_{Jump}**	**λ^2_{Jump}(Å²)**	**contribution of jump χ_{Jump}**	**Jump Frequency ν_{Jump} (ns⁻¹)**	**Percentage of Jump trajectory Segment** $P_{Jump} = \frac{}{n_w}$	**Viscosity (cp)**
x_P = 0.1											
225	0.009(5.97×10^{-4})	167.5	0.228	0.0035	0.0055	25673	8.097	38.89	0.257	4.351	300.95(88.041)
235	0.028(1.457×10^{-3})	60.3	0.236	0.0089	0.0191	65362	8.221	31.79	0.654	3.961	91.50(13.504)
245	0.068(4.507×10^{-3})	27.5	0.242	0.0169	0.0511	121804	8.344	24.85	1.218	3.355	37.80(5.350)
255	0.13 (4.314×10^{-3})	14.5	0.250	0.0288	0.1032	201908	8.579	21.82	2.019	2.930	17.52(0.663)
265	0.277(3.27×10^{-3})	7.4	0.257	0.0447	0.2323	305821	8.787	16.14	3.058	2.264	7.59(0.253)
275	0.467(1.07×10^{-2})	4.82	0.265	0.0582	0.4088	381142	9.158	12.46	3.811	1.830	4.40(0.292)
285	0.679(2.04×10^{-2})	3.34	0.267	0.0718	0.6072	469562	9.170	10.57	4.696	1.550	2.99(0.237)
300	1.173(2.08×10^{-2})	2.5	0.276	0.1046	1.0684	646832	9.699	8.92	6.468	1.617	1.78(0.053)
x_P = 0.2											
225	0.007(6.95×10^{-4})	209.2	0.241	0.003	0.004	19039	8.738	42.86	0.190	4.051	303.85(34.110)
235	0.017(1.06×10^{-4})	83.8	0.242	0.006	0.011	41727	8.571	35.29	0.417	3.506	126.69(25.984)
245	0.045(3.83×10^{-4})	33.1	0.247	0.013	0.032	91564	8.553	28.89	0.916	3.032	46.53(3.537)
255	0.090(1.21×10^{-3})	19.3	0.258	0.020	0.07	133379	9.110	22.22	1.334	2.575	21.66(2.335)
265	0.184(3.42×10^{-3})	11.0	0.265	0.032	0.152	205802	9.384	17.39	2.058	2.264	9.97(0.529)
275	0.324(1.04×10^{-2})	6.53	0.270	0.047	0.277	293636	9.572	14.51	2.936	1.909	5.84(0.290)
285	0.525(1.40×10^{-2})	3.93	0.274	0.064	0.461	396709	9.695	12.19	3.967	1.547	3.24(0.179)
300	0.842(2.29×10^{-2})	3.15	0.307	0.055	0.787	279198	11.843	6.53	2.792	0.866	2.17(0.203)
x_P = 0.5											
225	0.010(3.63×10^{-4})	170.0	0.283	0.0034	0.0066	18628	10.911	34.00	0.186	3.212	123.88(47.729)
235	0.020(7.98×10^{-4})	58.4	0.262	0.0058	0.0142	32263	10.844	29.00	0.323	2.581	60.88(17.566)
245	0.050(1.18×10^{-3})	27.0	0.267	0.0168	0.0332	107293	9.419	33.60	1.073	2.900	25.94(13.561)
255	0.083(3.07×10^{-3})	17.5	0.272	0.0229	0.0601	142204	9.683	27.59	1.422	2.490	16.02(4.516)
265	0.157(4.98×10^{-3})	10.85	0.278	0.0396	0.1174	236473	10.058	25.22	2.365	2.556	7.90(0.894)

275	0.227(4.38×10⁻³)	8.4	0.288	0.0468	0.1802	263297	10.657	20.62	2.633	2.213	6.01(0.597)
285	0.354(8.55×10⁻³)	5.0	0.278	0.0688	0.2852	416758	9.910	19.44	4.168	2.084	4.09(0.256)
300	0.624(1.96×10⁻²)	4.53	0.324	0.0635	0.5605	286966	13.283	10.18	2.870	1.291	2.11(0.084)
x_P = 0.8											
225	0.017(4.55×10⁻⁴)	94.2	0.300	0.0066	0.0104	32975	11.954	38.82	0.330	3.111	57.36(12.849)
235	0.031(1.25×10⁻³)	56.2	0.321	0.0082	0.0228	36347	13.471	26.45	0.363	2.054	33.77(4.621)
245	0.067(2.08×10⁻³)	31.17	0.339	0.0130	0.054	52786	14.764	19.40	0.5279	1.644	16.14(2.609)
255	0.121(4.38×10⁻³)	19.56	0.343	0.0235	0.0975	93817	15.019	19.42	0.938	1.832	8.56(0.629)
265	0.181(6.58×10⁻³)	17.5	0.355	0.0280	0.153	104318	16.088	15.47	1.043	1.827	6.31(0.756)
275	0.336(1.35×10⁻²)	9.62	0.366	0.0390	0.297	139154	16.837	11.61	1.392	1.337	3.37(0.255)
285	0.384(8.20×10⁻³)	8.0	0.358	0.0420	0.342	157486	16.005	10.94	1.575	1.260	2.88(0.116)
300	0.672(1.78×10⁻²)	6.153	0.380	0.0479	0.6241	160164	17.962	7.13	1.602	0.977	1.77(0.088)
x_P = 1.0											
225	0.031(9.78×10⁻⁴)	57.2	0.326	0.009	0.022	37848	13.829	29.03	0.378	2.175	25.81(1.738)
235	0.044(2.27×10⁻³)	45.88	0.332	0.011	0.033	45918	14.198	25.00	0.459	2.106	18.89(1.031)
245	0.099(1.48×10⁻³)	24.2	0.348	0.018	0.081	71202	15.366	18.18	0.712	1.724	9.20(0.278)
255	0.178(4.93×10⁻³)	15.6	0.355	0.029	0.149	110048	15.895	16.29	1.100	1.717	5.41(0.216)
265	0.274(4.51×10⁻³)	12.0	0.365	0.034	0.24	120757	16.687	12.41	1.208	1.450	3.90(0.051)
275	0.350(1.06×10⁻²)	9.80	0.365	0.038	0.312	139649	16.524	10.86	1.396	1.369	3.12(0.090)
285	0.539(1.38×10⁻²)	7.466	0.392	0.039	0.5	124602	18.856	7.24	1.246	0.922	2.11(0.081)
300	0.769(1.90×10⁻²)	5.90	0.396	0.053	0.716	165561	19.215	6.89	1.656	0.977	1.55(0.105)

Table S5. The results of water at different binary mixture compositions (x_P=0.0, 0.1, 0.2, 0.5, 0.8, and 1.0) diffusion coefficient D, t^*, r_2, jump-diffusion coefficient D_{Jump}, number of jump n_{Jump}, the percentage contribution of jump χ_{Jump}, jump frequency ν_{Jump}, and percentage of jump trajectory segment P_{jump}. The values in parenthesis are the standard errors of D.

Water (W/P)										
T(K)	**D /10⁻⁵ (cm²/sec)**	**t^* (x axis)**	**r_2 (nm)**	**D_{Jump} /10⁻⁵ (cm²/sec)**	**D_{Res} /10⁻⁵ (cm²/sec)**	**n_{Jump}**	**λ^2_{Jump}(Å²)**	**contribution of jump χ_{Jump}**	**Jump Frequency ν_{Jump} (ns⁻¹)**	**Percentage of Jump trajectory Segment $P_{Jump} = \frac{}{n_w}$**
x_P = 0.1										
225	0.009(6.27×10⁻⁴)	150	0.2215	0.0040	0.005	27810	7.706	44.44	0.309	4.682
235	0.029(1.57×10⁻³)	60.3	0.236	0.0090	0.02	65362	8.221	31.03	0.654	3.961
245	0.072(4.61×10⁻³)	20.0	0.2222	0.0185	0.0535	147427	7.537	25.69	1.474	3.397
255	0.138(4.52×10⁻³)	13.3	0.2455	0.0341	0.1039	221913	8.310	24.71	2.019	2.466
265	0.306(3.44×10⁻³)	6.8	0.2510	0.0486	0.2574	311265	8.428	15.88	3.458	2.353
275	0.486(1.12×10⁻²)	4.27	0.2567	0.0632	0.4228	396332	8.608	13.00	4.404	1.850
285	0.705(2.24×10⁻²)	3.38	0.2696	0.0721	0.6329	416634	9.348	10.23	4.629	1.528
300	1.222(2.28×10⁻²)	2.4	0.2725	0.113	1.2107	639578	9.502	9.2471	7.106	1.706
x_P = 0.2										
225	0.008(1.3×10⁻³)	180.0	0.2325	0.0032	0.0048	18212	8.334	40.00	0.228	4.139
235	0.019(1.20×10⁻³)	70.8	0.2329	0.0071	0.0119	42280	8.026	37.37	0.529	3.748
245	0.049(7.44×10⁻⁴)	30.2	0.2419	0.0149	0.0341	86077	8.283	30.41	1.076	3.251
255	0.098(1.46×10⁻³)	17.4	0.2519	0.0235	0.0745	129133	8.732	23.98	1.614	2.812
265	0.198(3.72×10⁻³)	9.75	0.2577	0.0372	0.1608	200845	8.891	18.79	2.511	2.437
275	0.346(1.14×10⁻²)	6.212	0.2650	0.0548	0.2912	283841	9.267	15.84	3.548	2.201
285	0.560(1.91×10⁻²)	4.27	0.2864	0.0580	0.502	263921	10.551	10.36	3.299	1.386
300	0.908(2.65×10⁻²)	2.9	0.2876	0.0851	0.8229	386945	10.562	9.37	4.837	1.403
x_P = 0.5										
225	0.010(4.6×10⁻⁴)	130.4	0.2549	0.0037	0.0063	11995	9.309	37.00	0.240	3.157
235	0.021(5.93×10⁻⁴)	61.5	0.2579	0.0069	0.0141	22183	9.301	32.86	0.444	2.739
245	0.053(1.4×10⁻³)	30.58	0.2686	0.0137	0.0393	41522	9.892	25.85	0.830	2.540
255	0.089(4.76×10⁻³)	19.13	0.2743	0.0201	0.0689	59443	10.140	22.58	1.189	2.273
265	0.166(5.61×10⁻³)	15.3	0.3041	0.0289	0.1371	70746	12.276	17.41	1.415	2.167
275	0.240(2.75×10⁻³)	8.7	0.2935	0.0408	0.1992	108280	11.299	17.00	2.166	1.885
285	0.387(1.99×10⁻²)	4.5	0.2738	0.0638	0.3232	195939	9.761	16.49	3.919	1.764
300	0.679(3.1×10⁻²)	4.274	0.3177	0.0718	0.6072	166033	12.981	10.57	3.321	1.395
x_P = 0.8										
225	0.015(1.79×10⁻³)	98.2	0.2813	0.0040	0.011	4512	10.880	26.67	0.226	2.234
235	0.031(2.36×10⁻³)	59.7	0.2953	0.0065	0.0245	6654	11.805	20.97	0.333	1.992

245	$0.062(4.59\times10^{-3})$	36.64	0.3236	0.0104	0.0516	8912	13.975	16.77	0.446	1.632
255	$0.112(5.01\times10^{-3})$	21.65	0.3262	0.0194	0.0926	16422	14.164	17.32	0.821	1.777
265	$0.166(8.0\times10^{-3})$	18.2	0.3396	0.0260	0.14	20283	15.398	15.66	1.014	1.847
275	$0.316(1.0\times10^{-2})$	10.2	0.3521	0.0371	0.2789	27417	16.249	11.74	1.371	1.399
285	$0.402(1.88\times10^{-2})$	8.5	0.3514	0.0449	0.3571	33487	16.089	11.17	1.674	1.424
300	$0.666(4.79\times10^{-2})$	6.0	0.368	0.0636	0.6024	43537	17.543	9.55	2.177	1.307

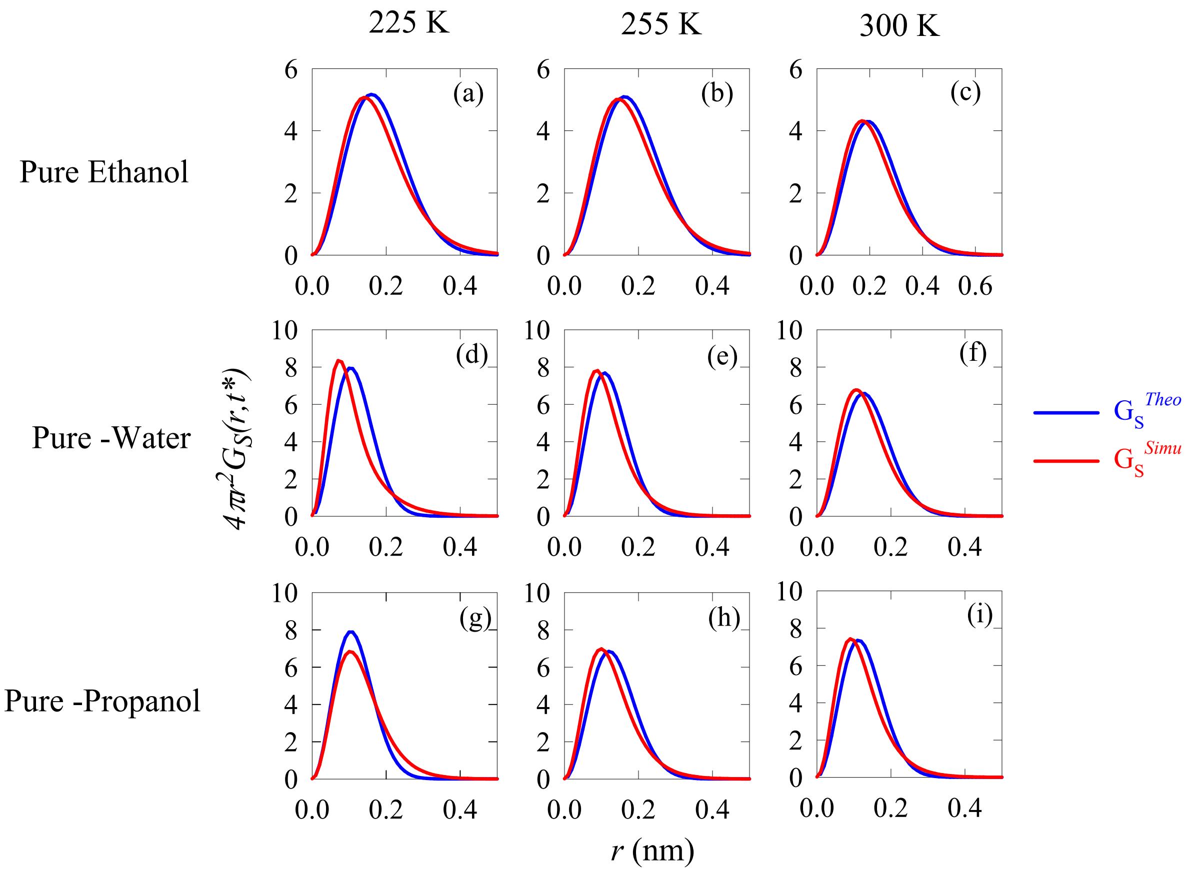

Figure S11. The simulated self-part of the van Hove correlation functions $G_S^{simu}(r,t^*)$ (red line) and the theoretical Gaussian distribution ($G_S^{theo}(r,t^*)$) (blue line) at time t^*at 225K, 255K and 300K temperatures for pure ethanol (panels: a, b, and c), pure water (panels: d, e, and f) and pure propanol (panels: g, h, and i). t^* is the time when $\alpha_2(t)$ is maximum.

W/E Mixture

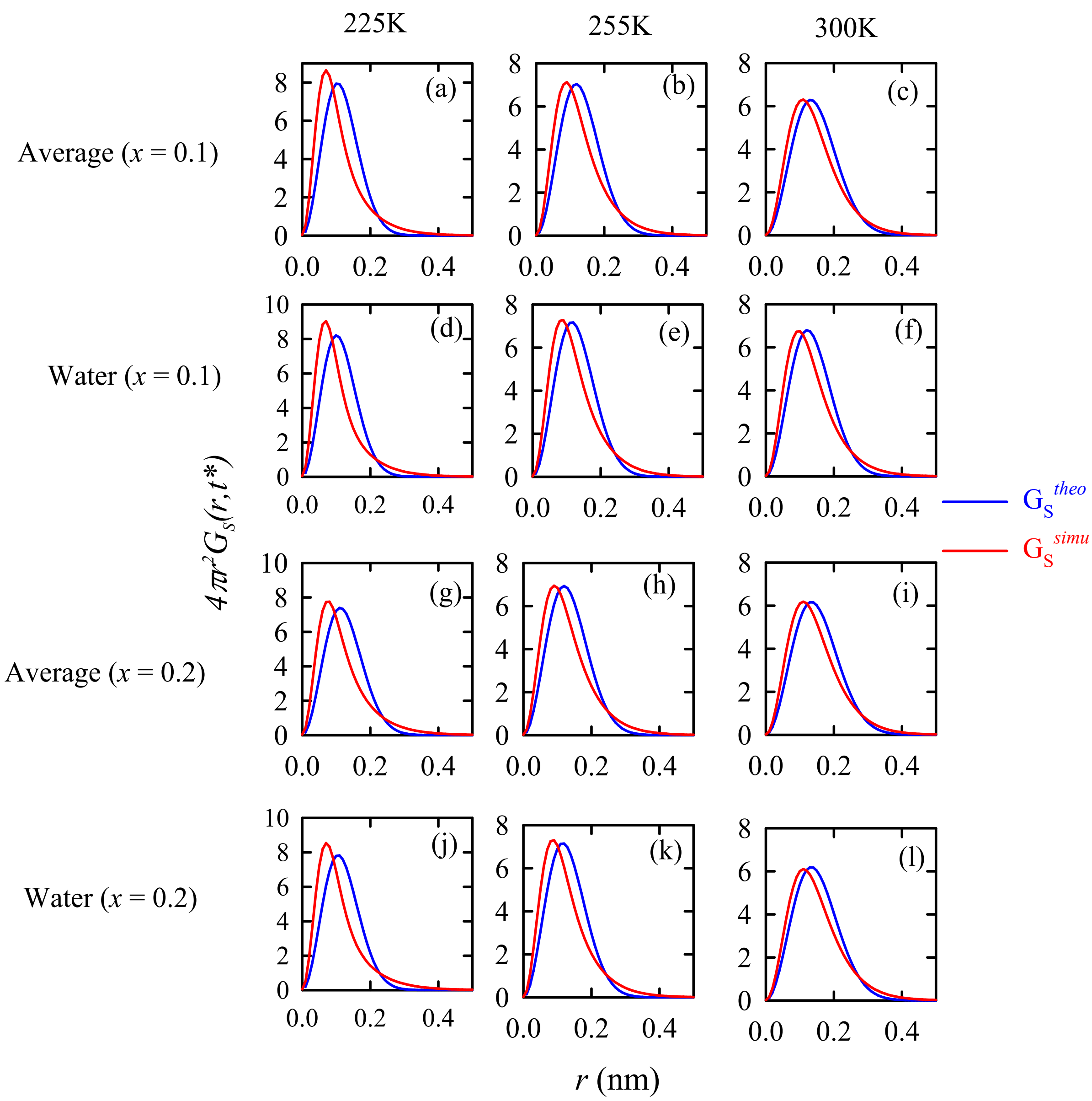

Figure S12. The simulated self-part of the van Hove correlation functions $G_S^{simu}(r,t^*)$ (red line) and the theoretical Gaussian distribution ($G_S^{theo}(r,t^*)$) (blue line) at time t^* for 225K, 255K, and 300K temperatures for the overall system and water for the W/E mixtures with ethanol mole fraction x_E=0 .1 and 0.2. t^* is the time when $\alpha_2(t)$ is maximum.

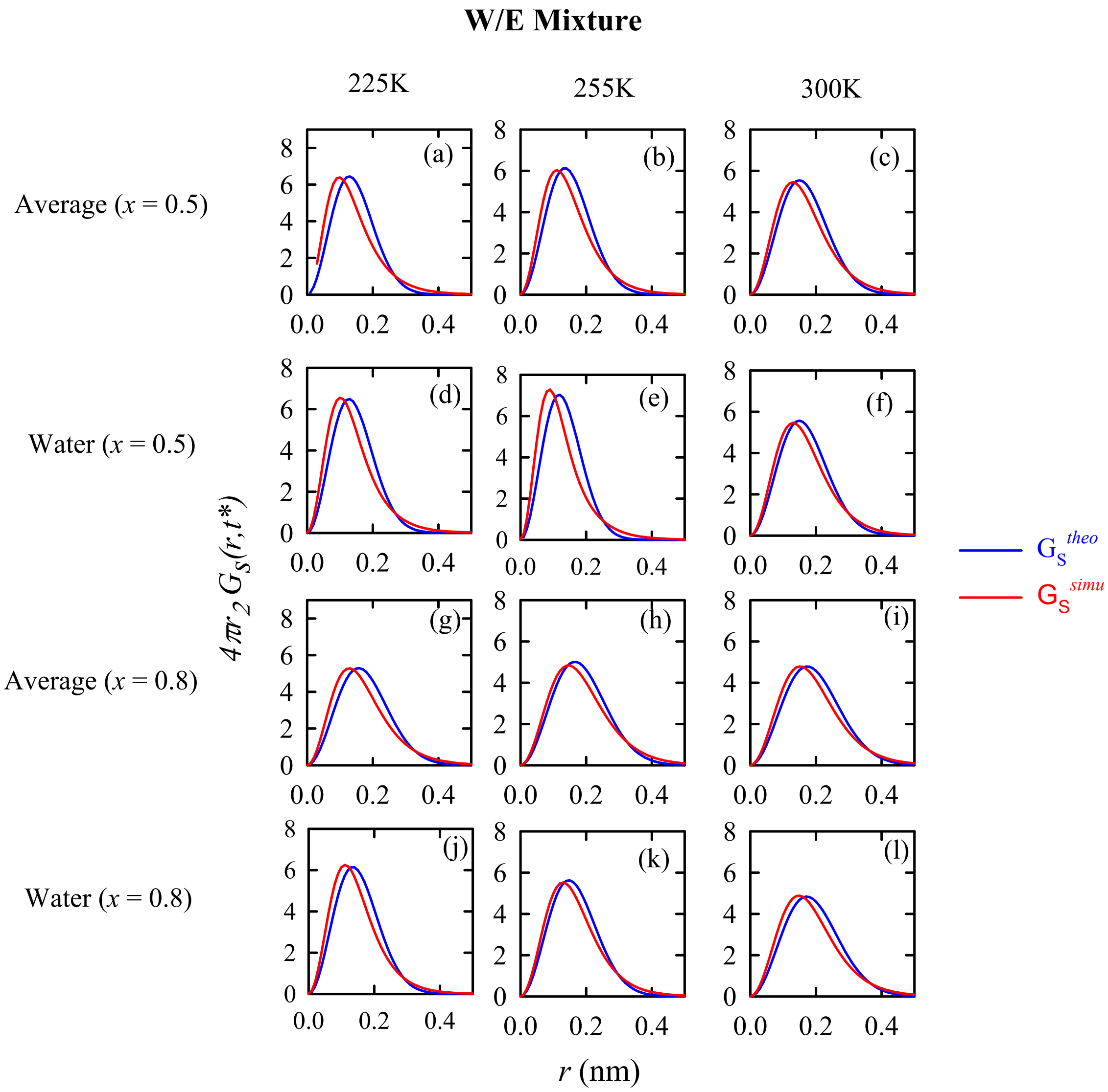

Figure S13. The simulated self-part of the van Hove correlation functions $G^{simu}_{S}(r,t^*)$ (red line) and the theoretical Gaussian distribution ($G^{theo}_{S}(r,t^*)$) (blue line) at time t^* for 225K, 255K, and 300K temperatures for the overall system and water for the W/E mixtures with ethanol mole fraction x_E=0 .5 and 0.8. t^* is the time when $\alpha_2(t)$ is maximum.

W/P Mixture

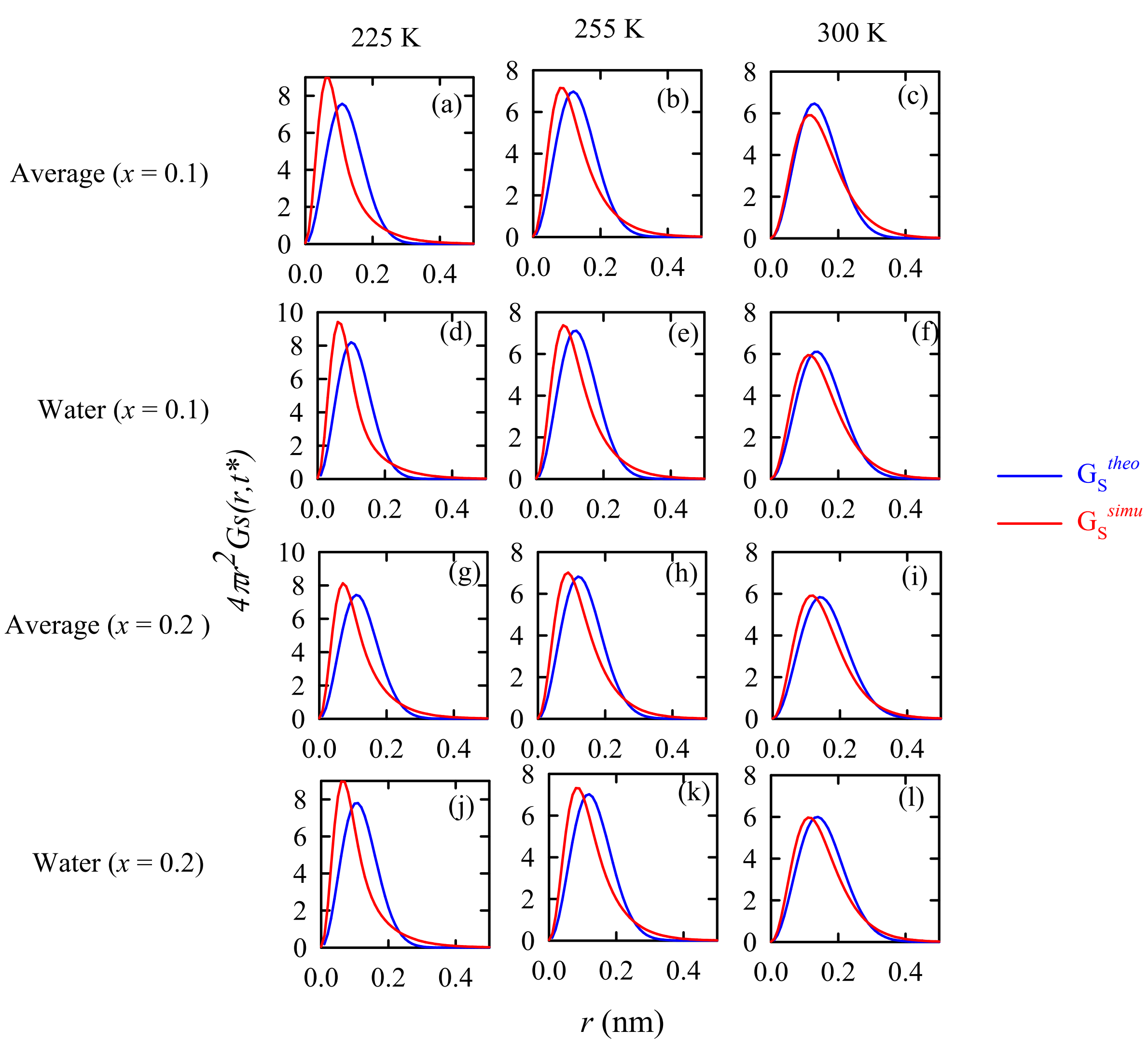

Figure S14. The simulated self-part of the van Hove correlation functions $G_S^{simu}(r,t^*)$ (red line) and the theoretical Gaussian distribution ($G_S^{theo}(r,t^*)$) (blue line) at time t^* for 225K, 255K, and 300K temperatures for the overall system and water for the W/P mixtures with 1-propanol mole fraction x_P=0 .1 and 0.2. t^* is the time when $\alpha_2(t)$ is maximum.

W/P Mixture

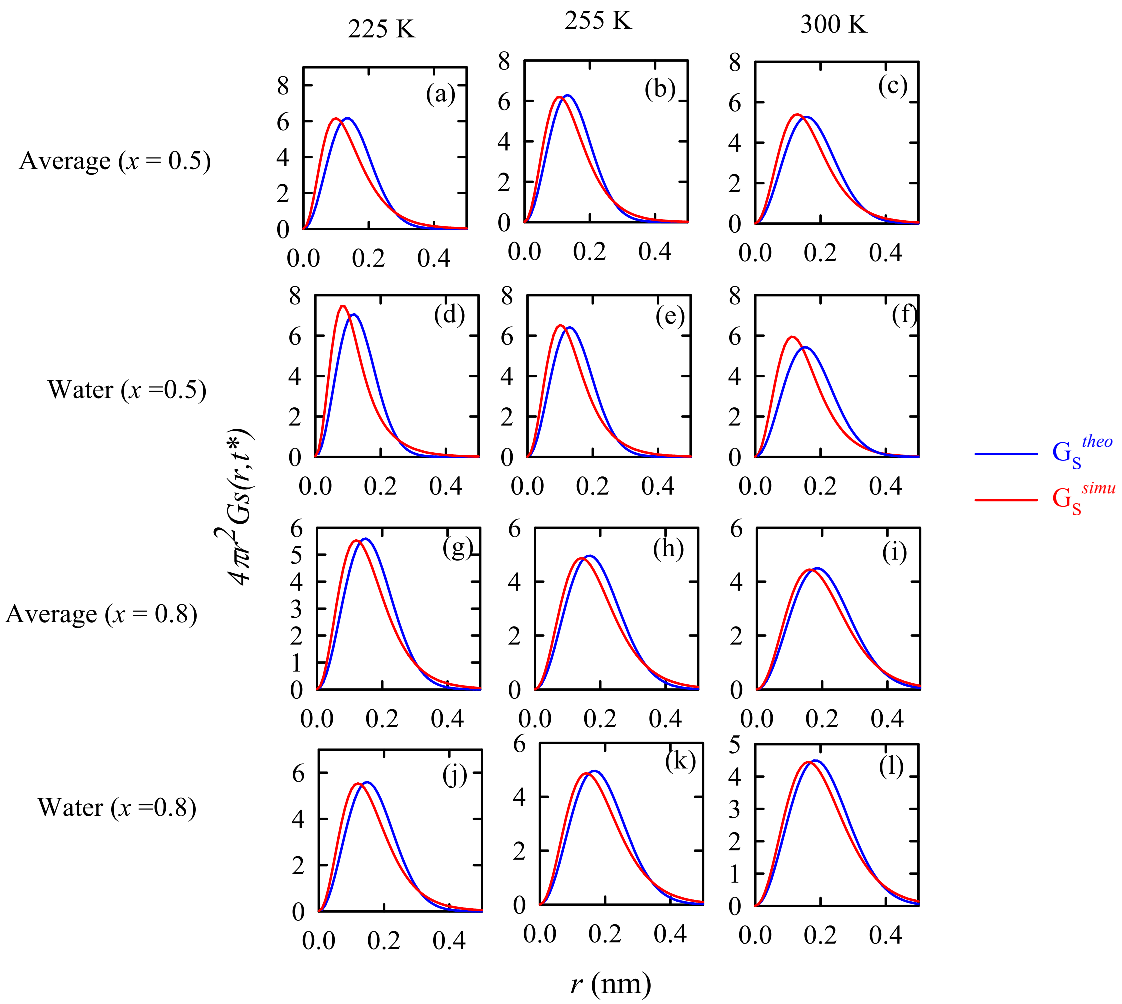

Figure S15. The simulated self-part of the van Hove correlation functions $G_S^{simu}(r,t^*)$ (red line) and the theoretical Gaussian distribution ($G_S^{theo}(r,t^*)$) (blue line) at time t^* for 225K, 255K, and 300K temperatures for the overall system and water for the W/P mixtures with ethanol mole fraction x_P=0 .5 and 0.8. t^* is the time when $\alpha_2(t)$ is maximum.

W/E Mixture

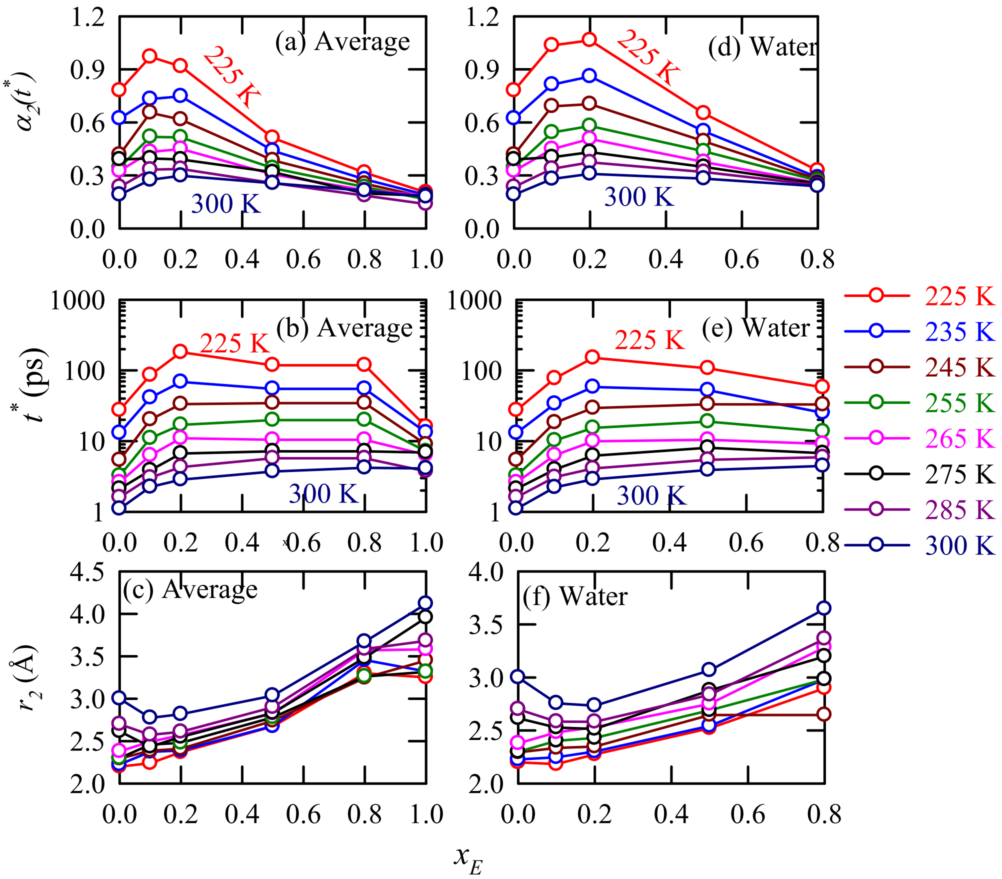

Figure S16. The height of $\alpha_2(t)$ peak, $\alpha_2(t^*)$, the characteristic time t^*, and the second crossing distance r_2 between $G_S^{simu}(r,t^*)$ and $G_S^{theo}(r,t^*)$ at different W/E mixture compositions and temperatures. The average values are presented in left panels, while the data for water is presented in the right-hand panels for W/E binary mixture.

W/P Mixture

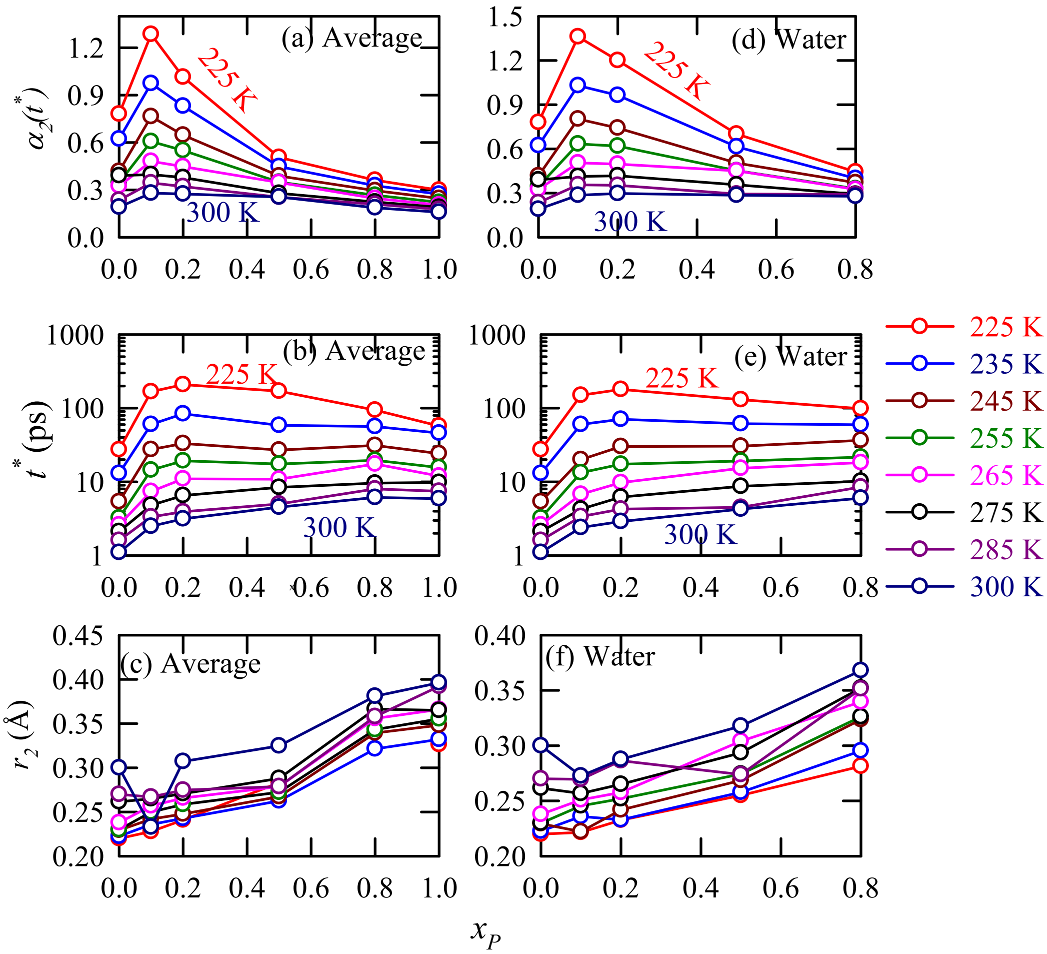

Figure S17. The height of $\alpha_2(t)$ peak, $\alpha_2(t^*)$, the characteristic time t^*, and the second crossing distance r_2 between $G_S^{simu}(r,t^*)$ and $G_S^{theo}(r,t^*)$ at different mixture compositions and temperatures. The average values are presented in left panels, while the data for water are presented in the right-hand panels for W/P binary mixture.

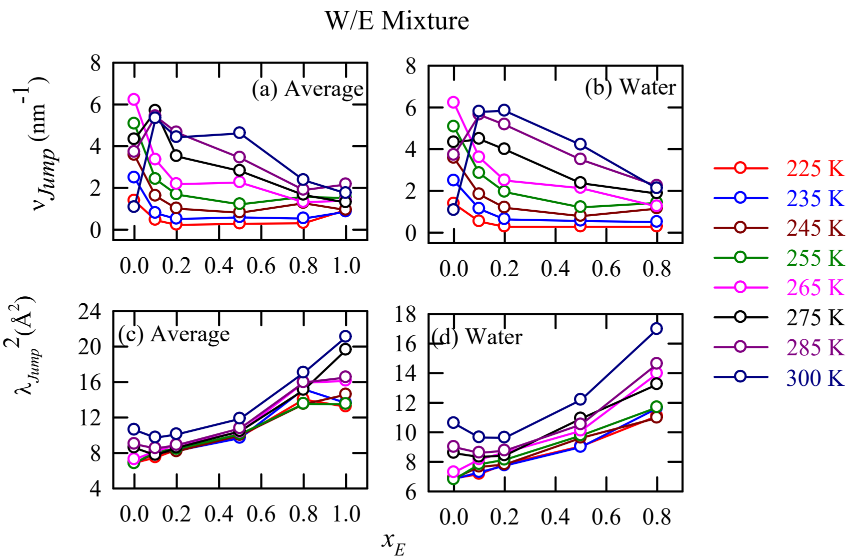

Figure S18. The composition-dependent ν_{Jump} and λ^{2}_{Jump} for different temperatures in W/E binary mixture.

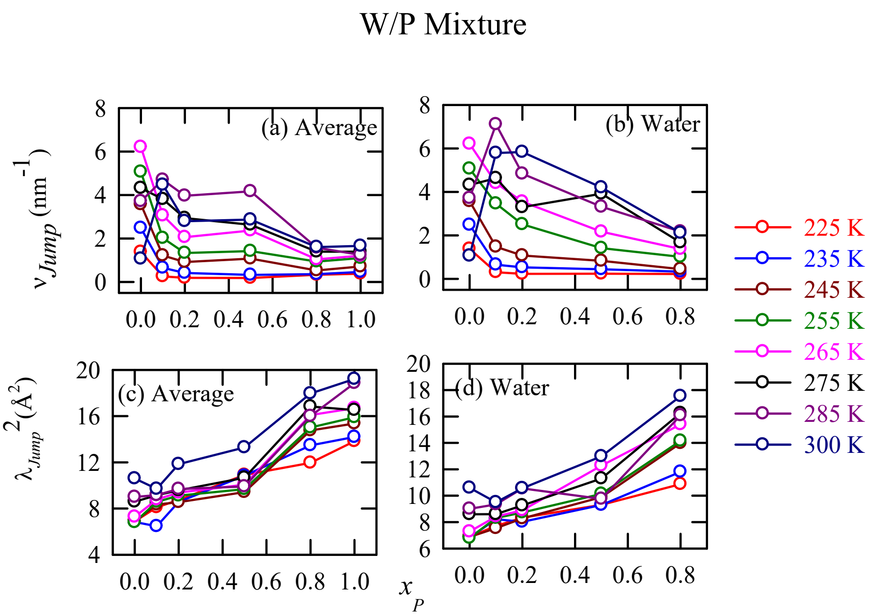

Figure S19. The composition-dependent ν_{Jump} and λ^{2}_{Jump} for different temperatures in W/P binary mixture.

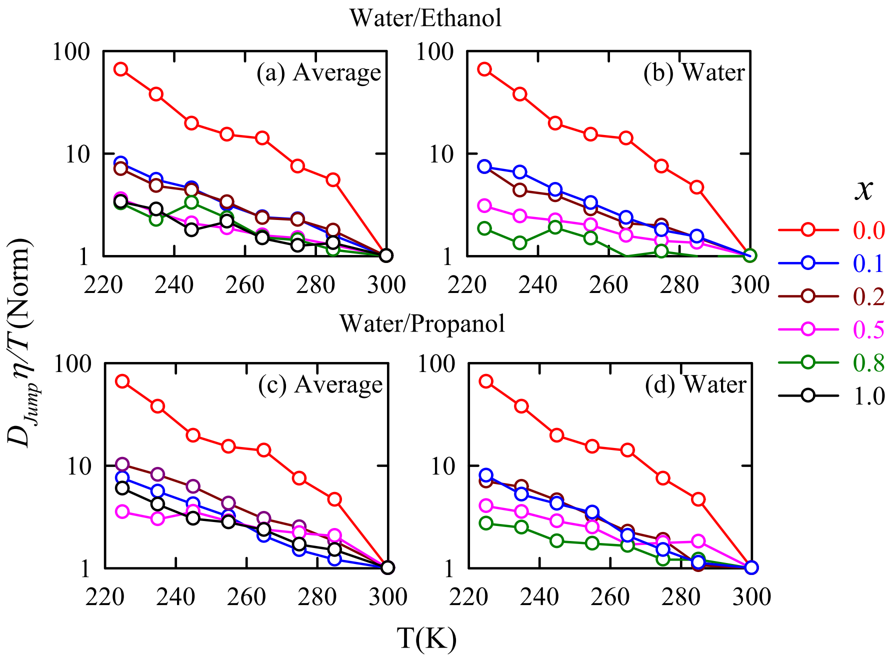

Figure S20. Temperature-dependent normalized $D_{Jump}\eta/T$ as a function of temperature. Average values and for water in two binary mixtures at different compositions. The normalizations are done at 300 K.

Table S6. The amplitudes and time-scales of H-bond correlations for the overall system at different temperatures and compositions of the W/E binary mixture.

average (W/E)								
	225K	**235K**	**245K**	**255K**	**265K**	**275K**	**285K**	**300K**
				x_E=0.0				
a_1	0.11	0.14	0.64	0.12	0.67	0.66	0.13	0.21
τ_1 (ps)	12.52	21.58	36.56	1.41	19.81	11.42	0.30	0.62
a_2	0.66	0.66	0.10	0.63	0.17	0.15	0.54	0.64
τ_2 (ps)	239.54	124.25	2.25	21.36	3.01	0.7	5.99	5.82
a_3	0.23	0.19	0.26	0.25	0.17	0.19	0.33	0.15
τ_3 (ps)	960.72	507.85	162.89	93.40	93.48	55.04	25.04	29.16
τ_{av} (ps)	380.44	181.52	65.97	36.98	29.68	18.10	11.54	8.23
				x_E=0.1				
a_1	0.08	0.26	0.12	0.18	0.11	0.15	0.15	0.26
τ_1 (ps)	18.35	111.21	15.02	15.08	0.91	2.81	1.13	2.73
a_2	0.57	0.60	0.61	0.63	0.56	0.63	0.56	0.59
τ_2 (ps)	672.15	456.82	146.29	84.81	30.53	27.18	14.79	14.39
a_3	0.35	0.14	0.27	0.19	0.33	0.22	0.29	0.15
τ_3 (ps)	2767.63	1968.59	567.10	377.14	148.29	117.13	66.50	66.70
τ_{av} (ps)	1353.26	578.61	243.59	127.80	66.13	43.31	27.74	19.21
				x_E=0.2				
a_1	0.06	0.17	0.10	0.09	0.10	0.11	0.13	0.16
τ_1 (ps)	14.83	118.47	17.63	1.65	1.21	0.96	0.74	1.05
a_2	0.42	0.61	0.47	0.52	0.55	0.57	0.56	0.56
τ_2 (ps)	800.17	657.91	186.86	83.92	50.81	32.91	21.11	13.65
a_3	0.51	0.22	0.44	0.39	0.35	0.32	0.31	0.28
τ_3 (ps)	3445.77	2726.84	703.41	401.60	254.50	162.96	103.72	61.91
τ_{av} (ps)	2094.30	1021.37	399.09	200.41	117.14	71.01	44.07	25.15
				x_E=0.5				
a_1	0.11	0.08	0.07	0.08	0.18	0.20	0.12	0.16
τ_1 (ps)	105.23	10.73	2.14	2.07	17.56	13.77	1.06	1.05
a_2	0.45	0.52	0.35	0.46	0.59	0.59	0.56	0.56

τ_2 (ps)	861.71	381.42	128.25	102.09	109.14	78.91	34.25	13.65
a_3	0.44	0.39	0.58	0.45	0.23	0.21	0.32	0.28
τ_3 (ps)	3450.04	1900.97	676.17	506.52	457.86	341.02	166.67	61.91
τ_{av} (ps)	1917.36	940.57	437.22	275.06	172.86	120.92	72.64	25.15
				x_E=0.8				
a_1	0.07	0.09	0.08	0.08	0.13	0.11	0.62	0.14
τ_1 (ps)	13.60	9.52	7.66	2.12	8.82	2.82	52.96	0.72
a_2	0.49	0.54	0.44	0.46	0.61	0.46	0.14	0.55
τ_2 (ps)	491.33	280.31	143.91	94.08	93.87	49.34	7.14	19.69
a_3	0.44	0.37	0.48	0.46	0.26	0.43	0.24	0.31
τ_3 (ps)	2278.85	1201.75	602.39	416.50	346.47	194.56	200.34	94.01
τ_{av} (ps)	1244.39	596.87	353.08	235.04	148.49	106.66	81.92	40.07
				x_E=1.0				
a_1	0.05	0.06	0.05	0.06	0.10	0.07	0.08	0.55
τ_1 (ps)	6.70	10.53	1.98	1.63	9.83	0.69	0.90	22.81
a_2	0.44	0.55	0.42	0.47	0.75	0.44	0.47	0.11
τ_2 (ps)	278.00	230.20	112.95	88.02	89.08	39.45	29.89	0.82
a_3	0.51	0.38	0.53	0.47	0.15	0.49	0.45	0.35
τ_3 (ps)	787.83	603.39	328.47	252.50	260.35	119.60	84.85	68.11
τ_{av} (ps)	524.45	356.57	221.63	160.14	106.84	76.01	52.30	36.48

Table S7. The amplitudes and time-scales of ethanol-ethanol H-bond correlation functions at different temperatures and compositions of the W/E binary mixture.

				ethanol-ethanol (W/E)				
	225K	235K	245K	255K	265K	275K	285K	300K
				x_E=0.1				
a_1	0.16	0.16	0.13	0.20	0.68	0.69	3.40	0.27
τ_1 (ps)	11.86	1.42	7.59	1.28	23.98	19.60	0.07	0.76
a_2	0.54	0.58	0.43	0.67	0.21	0.24	1.53	0.66
τ_2 (ps)	283.24	100.00	75.30	36.55	0.98	1.52	1.44	9.32
a_3	0.30	0.26	0.44	0.13	0.10	0.07	19.69	0.07
τ_3 (ps)	1491.25	660.51	359.22	258.19	203.23	129.88	14.71	83.40
τ_{av} (ps)	602.22	229.96	191.42	58.31	36.83	22.98	292.00	12.20
				x_E= 0.2				
a_1	0.15	0.33	0.09	0.25	0.27	0.60	0.11	0.35
τ_1 (ps)	14.72	93.34	1.64	11.67	7.60	23.80	1.22	3.31
a_2	0.48	0.54	0.44	0.66	0.63	0.22	0.51	0.57
τ_2 (ps)	348.13	251.57	110.57	79.67	53.81	1.67	30.22	16.63
a_3	0.38	0.13	0.46	0.10	0.10	0.18	0.38	0.08
τ_3 (ps)	1722.05	2320.06	563.22	453.25	521.14	130.17	98.00	144.16
τ_{av} (ps)	823.69	468.26	307.88	100.82	88.07	38.08	52.79	22.17
				x_E= 0.5				
a_1	0.20	0.15	0.10	0.14	0.14	0.16	0.19	0.19
τ_1 (ps)	87.50	12.38	18.71	1.62	2.26	1.79	2.62	1.35
a_2	0.62	0.43	0.39	0.53	0.50	0.56	0.63	0.64
τ_2 (ps)	669.61	188.54	189.37	70.73	42.66	34.21	31.52	19.66
a_3	0.18	0.42	0.50	0.34	0.35	0.29	0.19	0.17
τ_3 (ps)	2882.61	801.73	731.21	306.14	175.64	141.41	121.25	78.72
τ_{av} (ps)	951.53	419.65	441.33	141.80	83.12	60.46	43.39	26.22
				x_E= 0.8				
a_1	0.09	0.45	0.26	0.12	0.09	0.10	0.11	0.14
τ_1 (ps)	11.15	171.70	106.79	17.32	1.00	1.65	1.22	1.60
a_2	0.59	0.08	0.34	0.68	0.44	0.48	0.51	0.64
τ_2 (ps)	456.86	3.95	562.32	135.59	45.87	38.98	30.22	24.88
a_3	0.32	0.47	0.40	0.13	0.47	0.42	0.38	0.23
τ_3 (ps)	1508.30	597.88	808.97	458.63	161.79	129.23	98.00	76.73
τ_{av} (ps)	753.20	358.58	542.54	153.90	96.31	73.15	52.79	33.79

Table S8. The amplitudes and time-scales of water-water H-bond correlations at different temperatures and compositions of the W/E binary mixture.

water-water (W/E)								
	225K	**235K**	**245K**	**255K**	**265K**	**275K**	**285K**	**300K**
				x_E=0.1				
a_1	0.07	0.08	0.12	0.18	0.16	0.15	0.14	0.18
τ_1 (ps)	38.87	8.30	15.02	15.44	2.31	3.02	1.15	0.94
a_2	0.51	0.59	0.61	0.62	0.48	0.62	0.56	0.61
τ_2 (ps)	656.05	255.42	146.29	86.31	29.38	27.66	14.89	10.74
a_3	0.42	0.33	0.27	0.20	0.40	0.23	0.30	0.21
τ_3 (ps)	2565.95	1126.79	567.10	374.20	131.57	117.74	66.97	48.24
τ_{av} (ps)	1415.00	523.20	244.15	131.13	67.10	44.68	28.59	16.85
				x_E= 0.2				
a_1	0.05	0.17	0.12	0.08	0.16	0.11	0.12	0.15
τ_1 (ps)	15.35	75.00	15.02	2.51	4.18	1.01	0.75	1.04
a_2	0.38	0.36	0.61	0.58	0.56	0.57	0.56	0.56
τ_2 (ps)	827.86	817.03	146.29	104.41	65.01	34.47	21.32	13.73
a_3	0.57	0.47	0.27	0.34	0.31	0.33	0.32	0.29
τ_3 (ps)	3486.62	875.94	567.10	470.86	285.99	167.58	103.51	62.54
τ_{av} (ps)	2302.73	718.57	244.15	220.85	125.73	75.06	45.15	25.98
				x_E= 0.5				
a_1	0.06	0.07	0.37	0.06	0.16	0.10	0.09	0.32
τ_1 (ps)	2.28	20.69	196.53	2.28	2.60	4.71	0.97	11.08
a_2	0.49	0.61	0.28	0.49	0.57	0.59	0.56	0.53
τ_2 (ps)	132.56	591.54	797.41	132.56	85.35	63.25	36.24	45.28
a_3	0.45	0.32	0.28	0.45	0.35	0.31	0.35	0.15
τ_3 (ps)	615.33	2640.82	797.38	615.33	406.60	297.67	184.33	192.53
τ_{av} (ps)	341.99	1207.35	519.25	341.99	191.38	130.07	84.90	56.43
				x_E= 0.8				
a_1	0.06	0.41	0.08	0.07	0.16	0.11	0.08	0.18
τ_1 (ps)	52.65	814.57	7.66	9.44	5.61	7.56	3.11	11.09
a_2	0.44	0.50	0.44	0.45	0.43	0.46	0.48	0.58
τ_2 (ps)	1013.91	819.16	143.91	178.55	92.91	80.29	51.59	52.74
a_3	0.51	0.09	0.48	0.48	0.50	0.44	0.45	0.24
τ_3 (ps)	3191.17	45.54	602.39	627.85	379.27	309.53	211.08	181.41
τ_{av} (ps)	2076.77	747.65	353.08	382.38	230.48	173.96	120.00	76.12

Table S9. The amplitudes and time scales of water-ethanol H-bond correlations at different temperatures and compositions of the W/E binary mixture.

water-ethanol (W/E)								
	225K	**235K**	**245K**	**255K**	**265K**	**275K**	**285K**	**300K**
				x_E= 0.1				
a_1	0.20	0.16	0.13	0.12	0.16	0.15	0.69	0.64
τ_1 (ps)	126.61	16.95	7.58	1.05	2.14	0.69	7.23	10.75
a_2	0.63	0.53	0.43	0.60	0.60	0.59	0.05	0.20
τ_2 (ps)	780.88	198.54	75.30	46.53	33.23	19.57	0.04	0.63
a_3	0.17	0.31	0.44	0.28	0.25	0.26	0.26	0.16
τ_3 (ps)	379.27	952.13	359.22	236.11	157.35	95.59	85.47	54.37
τ_{av} (ps)	582.30	402.11	190.60	93.85	59.07	36.83	27.19	15.75
				x_E= 0.2				
a_1	0.08	0.21	0.09	0.11	0.16	0.14	0.15	0.20
τ_1 (ps)	7.61	46.53	1.64	1.30	0.96	0.83	1.32	1.53
a_2	0.42	0.36	0.44	0.51	0.53	0.56	0.54	0.61
τ_2 (ps)	546.44	720.88	110.57	67.07	42.65	29.46	20.47	16.13
a_3	0.50	0.42	0.46	0.39	0.35	0.30	0.31	0.19
τ_3 (ps)	2697.10	622.94	563.22	622.94	221.95	147.55	90.39	70.33
τ_{av} (ps)	1589.54	537.06	310.34	275.14	99.66	60.98	39.10	23.70
				x_E= 0.5				
a_1	0.07	0.08	0.10	0.09	0.16	0.11	0.12	0.14
τ_1 (ps)	12.29	7.11	18.71	2.39	1.39	1.04	1.95	0.79
a_2	0.40	0.46	0.39	0.45	0.47	0.48	0.50	0.54
τ_2 (ps)	552.31	313.84	189.37	104.66	56.95	40.36	32.78	20.50
a_3	0.53	0.46	0.50	0.46	0.43	0.41	0.38	0.32
τ_3 (ps)	2877.89	1623.08	731.21	487.11	283.15	200.69	145.13	90.27

τ_{av} (ps)	1755.74	894.38	445.60	270.25	148.94	102.36	71.47	39.93
				x_E= 0.8				
a_1	0.08	0.22	0.26	0.07	0.16	0.09	0.10	0.14
τ_1 (ps)	26.64	81.32	106.79	3.02	2.45	1.77	1.75	4.85
a_2	0.41	0.42	0.34	0.37	0.39	0.46	0.44	0.55
τ_2 (ps)	694.79	841.92	562.32	109.14	71.46	60.04	41.67	38.60
a_3	0.51	0.37	0.40	0.56	0.53	0.46	0.46	0.31
τ_3 (ps)	2734.98	759.07	808.97	475.72	475.72	235.79	162.52	124.34
τ_{av} (ps)	1680.40	647.91	542.83	306.12	278.62	134.98	93.47	60.25

Table S10. The amplitudes and time scales for the H-bond correlations for the overall system at different temperatures and different compositions of the W/P binary mixture.

				average (W/P)				
	225K	**235K**	**245K**	**255K**	**265K**	**275K**	**285K**	**300K**
				x_P= 0.0				
a_1	0.11	0.14	0.64	0.12	0.67	0.66	0.13	0.21
τ_1 (ps)	12.52	21.58	36.56	1.41	19.81	11.42	0.30	0.62
a_2	0.66	0.66	0.10	0.63	0.17	0.15	0.54	0.64
τ_2 (ps)	239.54	124.25	2.25	21.36	3.01	0.7	5.99	5.82
a_3	0.23	0.19	0.26	0.25	0.17	0.19	0.33	0.15
τ_3 (ps)	960.72	507.85	162.89	93.40	93.48	55.04	25.04	29.16
τ_{av} (ps)	380.44	181.52	65.97	36.98	29.68	18.10	11.54	8.23
				x_P= 0.1				
a_1	0.25	0.33	0.31	0.10	0.11	0.13	0.18	0.18
τ_1 (ps)	3058.12	23897.93	833.75	17.98	9.49	15.90	19.92	6.16
a_2	0.32	0.33	0.55	0.56	0.56	0.57	0.61	0.58
τ_2 (ps)	42395.54	26553.25	3493.24	697.50	363.08	240.02	189.62	97.29
a_3	0.42	0.33	0.14	0.34	0.33	0.30	0.22	0.24
τ_3 (ps)	21220.04	29208.58	14816.08	3618.26	1805.28	1120.62	847.40	494.21
τ_{av} (ps)	2347.74	2660.64	427.06	162.40	80.62	47.26	30.38	17.64
				x_P= 0.2				
a_1	0.05	0.09	0.08	0.10	0.11	0.35	0.35	0.17
τ_1 (ps)	723.24	362.25	31.88	24.35	12.55	168.10	168.10	6.48
a_2	0.31	0.52	0.51	0.57	0.56	0.52	0.52	0.58
τ_2 (ps)	8629.08	5177.12	1764.30	973.73	505.96	717.77	717.77	129.06
a_3	0.59	0.39	0.41	0.34	0.34	0.13	0.13	0.26
τ_3 (ps)	39038.21	24703.02	8935.30	5243.56	2816.22	3476.73	3476.73	698.14
τ_{av} (ps)	2564.38	1229.12	458.68	231.70	122.95	88.71	88.71	25.47
				x_P= 0.5				
a_1	0.31	0.27	0.10	0.09	0.10	0.13	0.14	0.24
τ_1 (ps)	4462.03	4523.34	138.15	29.93	17.50	46.26	29.68	65.23
a_2	0.03	0.25	0.53	0.53	0.55	0.58	0.57	0.57
τ_2 (ps)	777.47	1890.94	2329.14	1347.40	765.10	642.18	425.88	379.94
a_3	0.65	0.49	0.38	0.37	0.35	0.29	0.29	0.18
τ_3 (ps)	33364.03	23607.36	11872.74	7810.73	4543.93	3493.58	2330.66	1965.38
τ_{av} (ps)	2326.92	1317.40	570.40	362.64	199.96	139.83	92.73	59.36
				x_P= 0.8				
a_1	0.09	0.08	0.07	0.08	0.09	0.26	0.24	0.12
τ_1 (ps)	147.22	155.32	41.14	27.03	23.29	323.96	220.15	6.87
a_2	0.55	0.53	0.44	0.51	0.55	0.58	0.60	0.52
τ_2 (ps)	36782.55	6054.05	2404.56	1517.18	1184.71	1384.13	1016.70	307.02
a_3	0.35	0.39	0.48	0.41	0.35	0.16	0.16	0.35
τ_3 (ps)	8339.72	27609.36	11585.25	7382.83	5620.18	5405.14	4186.58	1439.63
τ_{av} (ps)	2331.16	1396.10	668.54	381.23	264.64	175.73	133.93	67.09
				x_P= 1.0				
a_1	0.06	0.06	0.05	0.16	0.12	0.10	0.10	0.11
τ_1 (ps)	485.83	122.00	24.83	662.20	293.59	121.33	66.06	17.32
a_2	0.43	0.58	0.42	0.70	0.69	0.67	0.67	0.62
τ_2 (ps)	8992.15	6506.77	2408.02	2920.06	1702.04	1121.77	712.08	409.80
a_3	0.50	0.36	0.53	0.14	0.20	0.23	0.23	0.27
τ_3 (ps)	26379.39	20844.81	8512.81	10050.39	5310.14	3631.37	2326.86	1271.34
τ_{av} (ps)	1719.60	1131.16	550.27	356.64	224.09	158.73	101.89	59.55

Table S11. The amplitudes and time-scales of water-water H-bond correlations at different temperatures and compositions of the W/P binary mixture.

water-water (W/P)								
x_P= 0.1								
	225K	235K	245K	255K	265K	275K	285K	300K
a_1	0.07	0.09	0.14	0.10	0.10	0.13	0.59	0.59
τ_1 (ps)	729.70	172.83	146.29	18.81	9.90	13.01	158.98	102.90
a_2	0.55	0.59	0.55	0.56	0.56	0.53	0.15	0.18
τ_2 (ps)	38740.03	4336.71	1925.95	689.80	363.93	213.21	8.23	8.90
a_3	0.38	0.33	0.31	0.35	0.34	0.34	0.26	0.23
τ_3 (ps)	9190.00	19842.98	8425.48	3579.2	1810.09	1024.96	802.55	521.38
τ_{av} (ps)	2475.00	901.18	369.37	163.37	81.50	46.64	30.26	18.05
x_P= 0.2								
a_1	0.05	0.07	0.09	0.12	0.35	0.12	0.15	0.53
τ_1 (ps)	103.67	115.32	63.32	95.19	329.54	8.93	18.09	240.75
a_2	0.31	0.47	0.56	0.58	0.47	0.56	0.60	0.33
τ_2 (ps)	7282.03	4258.63	2198.64	1165.94	1361.00	289.26	222.57	51.76
a_3	0.64	0.46	0.35	0.30	0.09	0.33	0.25	0.14
τ_3 (ps)	38953.63	22473.61	10306.36	5807.34	6478.42	1611.26	1197.21	1236.66
τ_{av} (ps)	2720.42	1233.29	484.05	242.75	131.49	68.74	43.57	31.25
x_P= 0.5								
a_1	0.05	0.13	0.20	0.17	0.13	0.19	0.14	0.14
τ_1 (ps)	133.24	1372.50	884.98	323.39	106.11	102.17	44.90	11.60
a_2	0.37	0.57	0.40	0.47	0.61	0.50	0.60	0.58
τ_2 (ps)	7955.98	7959.67	2354.20	1590.88	1082.17	644.46	473.33	234.15
a_3	0.58	0.30	0.40	0.36	0.26	0.32	0.26	0.28
τ_3 (ps)	38640.53	36215.25	12342.52	8879.13	6746.96	3747.73	2916.96	1531.03
τ_{av} (ps)	2529.76	1555.10	609.29	402.13	241.14	152.55	104.54	57.03
x_P= 0.8								
a_1	0.30	0.05	0.05	0.22	0.19	0.17	0.34	0.35
τ_1 (ps)	5567.04	198.29	120.19	718.69	339.10	159.86	359.98	214.24
a_2	0.36	0.43	0.40	0.38	0.41	0.43	0.45	0.42
τ_2 (ps)	36739.34	5892.09	2694.11	2175.74	1540.96	846.99	1394.59	844.04
a_3	0.34	0.52	0.54	0.40	0.40	0.41	0.21	0.23
τ_3 (ps)	47168.79	31548.47	14558.19	10983.33	7632.03	4528.12	5287.35	3015.09
τ_{av} (ps)	3105.91	1886.85	900.91	539.16	372.84	222.52	186.00	112.57

Table S12. The amplitudes and time-scales of 1-propanol-1-propanol H-bond correlations at different temperatures and compositions of the W/P binary mixture.

1-propanol-1-propanol (W/P)								
x_P= 0.1								
	225K	235K	245K	255K	265K	275K	285K	300K
a_1	0.05	0.14	0.16	0.16	0.19	0.71	0.14	0.26
τ_1 (ps)	167.55	9.17	105.00	16.78	16.97	332.79	772.67	5.78
a_2	0.41	0.49	0.66	0.55	0.58	0.24	0.23	0.64
τ_2 (ps)	3065.66	1676.20	1540.18	561.35	303.51	26.17	6.90	111.93
a_3	0.54	0.37	0.18	0.29	0.23	0.05	0.63	0.09
τ_3 (ps)	19803.65	8756.85	7087.79	2605.91	1294.57	1538.86	158.27	724.00
τ_{av} (ps)	1200.92	405.11	231.04	106.60	47.68	32.62	20.97	14.12
x_P= 0.2								
a_1	0.12	0.12	0.18	0.15	0.28	0.60	0.20	0.28
τ_1 (ps)	399.25	98.87	160.46	22.75	125.77	305.52	10.65	30.29
a_2	0.35	0.49	0.64	0.47	0.59	0.19	0.61	0.61
τ_2 (ps)	5659.30	2816.51	1846.06	621.59	730.29	13.72	215.48	203.80
a_3	0.53	0.39	0.18	0.39	0.13	0.22	0.18	0.10
τ_3 (ps)	20490.16	13295.8	9562.07	2839.46	4020.14	1675.21	1223.58	1541.84
τ_{av} (ps)	1291.51	656.80	297.60	138.85	99.17	54.66	35.94	29.43
x_P= 0.5								
a_1	0.09	0.21	0.31	0.12	0.62	0.15	0.16	0.18
τ_1 (ps)	65.38	943.69	1302.81	17.02	1212.32	22.94	14.84	9.96

a₂	0.40	0.59	0.17	0.52	0.23	0.54	0.53	0.58
τ_2 (ps)	5097.09	6913.76	198.85	1074.56	180.20	486.93	320.16	217.10
a_3	0.51	0.20	0.52	0.36	0.15	0.31	0.32	0.24
τ_3 (ps)	29412.38	30448.9	6707.54	5405.10	6082.06	2183.36	1461.92	985.60
τ_{av} (ps)	1702.53	1040.29	389.78	248.19	171.12	94.44	63.19	36.60
x_P= 0.8								
a_1	0.07	0.08	0.11	0.20	0.19	0.11	0.11	0.21
τ_1 (ps)	111.29	96.43	139.83	530.07	322.12	9.80	16.64	92.05
a_2	0.38	0.55	0.45	0.66	0.67	0.55	0.52	0.67
τ_2 (ps)	6667.84	5188.03	2569.25	2641.01	1766.05	591.64	455.08	498.73
a_3	0.55	0.37	0.44	0.14	0.14	0.34	0.37	0.12
τ_3 (ps)	27188.89	20155.72	8873.23	10723.94	7350.03	2296.06	1713.91	2360.1
τ_{av} (ps)	1757.11	1023.71	509.12	334.97	227.94	111.21	87.22	63.40

Table S13. The amplitudes and time-scales of water-1-propanol H-bond correlations at different temperatures and compositions of the W/P binary mixture.

water-1-propanol (W/P)								
x_P= 0.1								
	225K	**235K**	**245K**	**255K**	**265K**	**275K**	**285K**	**300K**
a_1	0.08	0.09	0.10	0.36	0.20	0.30	0.30	0.22
τ_1 (ps)	268.53	50.17	23.11	432.26	90.99	107.25	107.25	18.38
a_2	0.40	0.58	0.58	0.53	0.64	0.58	0.58	0.61
τ_2 (ps)	6681.68	3467.90	1471.75	1734.79	599.39	480.07	480.07	147.16
a_3	0.52	0.33	0.32	0.11	0.17	0.12	0.12	0.17
τ_3 (ps)	30356.00	16780.39	7743.96	9069.86	3012.12	2467.88	2467.88	685.32
τ_{av} (ps)	1860.43	758.93	334.27	207.65	90.04	61.20	61.20	21.08
x_P= 0.2								
a_1	0.07	0.07	0.09	0.11	0.12	0.13	0.15	0.18
τ_1 (ps)	239.00	34.59	20.31	15.46	9.13	7.36	6.59	9.44
a_2	0.61	0.46	0.50	0.54	0.52	0.52	0.54	0.56
τ_2 (ps)	34760.27	3372.67	1588.59	911.27	477.39	297.18	201.63	149.58
a_3	0.32	0.47	0.40	0.35	0.36	0.34	0.31	0.26
τ_3 (ps)	6365.40	19696.19	8528.42	5080.35	2649.11	1550.16	1033.54	721.47
τ_{av} (ps)	2316.99	1077.63	425.03	227.45	120.75	68.94	43.03	27.34
x_P= 0.5								
a_1	0.15	0.09	0.16	0.09	0.11	0.22	0.13	0.15
τ_1 (ps)	1565.65	145.25	542.12	20.55	18.36	158.38	24.91	14.96
a_2	0.21	0.50	0.52	0.46	0.52	0.57	0.53	0.54
τ_2 (ps)	6184.76	5091.09	3251.18	1151.26	777.41	970.97	406.37	245.24
a_3	0.64	0.41	0.32	0.45	0.37	0.22	0.34	0.31
τ_3 (ps)	33852.93	26742.63	13474.92	6885.68	4311.52	4266.18	2072.11	1205.31
τ_{av} (ps)	2319.34	1355.98	609.42	361.12	199.78	150.86	91.69	51.30
x_P= 0.8								
a_1	0.25	0.07	0.07	0.08	0.23	0.10	0.10	0.57
τ_1 (ps)	3931.29	192.95	60.72	39.95	618.84	19.80	14.41	771.66
a_2	0.35	0.43	0.39	0.44	0.52	0.49	0.51	0.26
τ_2 (ps)	34297.94	5965.73	2602.69	1610.71	2887.60	773.95	597.15	161.57
a_3	0.40	0.50	0.54	0.48	0.17	0.41	0.39	0.17
τ_3 (ps)	40109.53	28877.24	13134.65	8112.73	9662.73	3474.87	2707.71	2798.2
τ_{av} (ps)	2890.31	1696.03	812.55	463.56	325.13	181.28	136.39	95.14

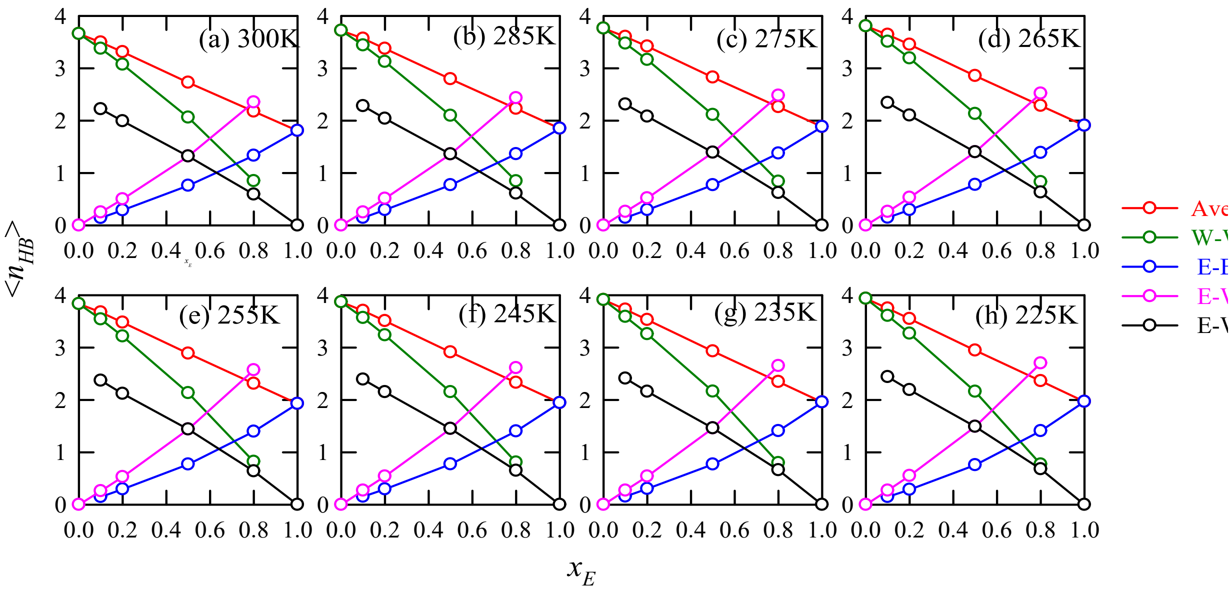

Figure S21. The number of water-water H-bonds per water, ethanol-ethanol H-bonds per ethanol molecule, and the number of ethanol-water H-bonds per ethanol and per water molecule, and average number of H-bonds in W/E binary mixture for x_E = 0.0, 0.1, 0.2, 0.5, 0.8, and 1.0.

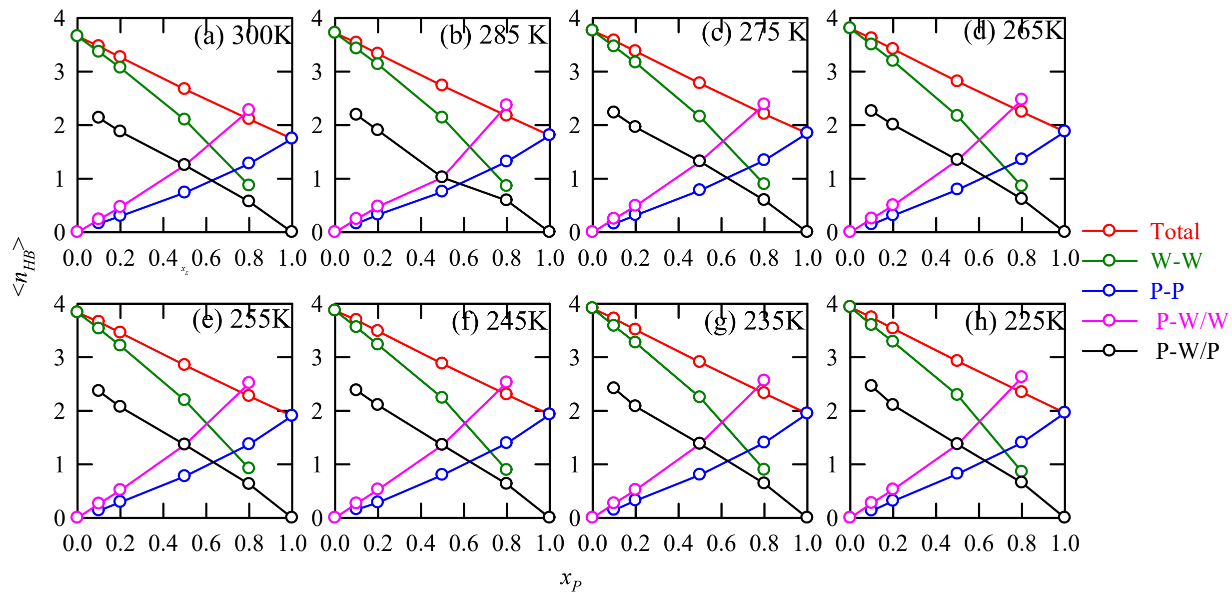

Figure S22. The number of water-water H-bonds per water, 1-propanol-1-propanol H-bonds per 1-propanol molecule, and the number of 1-propanol-water H-bonds per 1-proapnol and per water molecule, and average number of H-bonds in W/P binary mixture for x_P = 0.0, 0.1, 0.2, 0.5, 0.8, and 1.0.

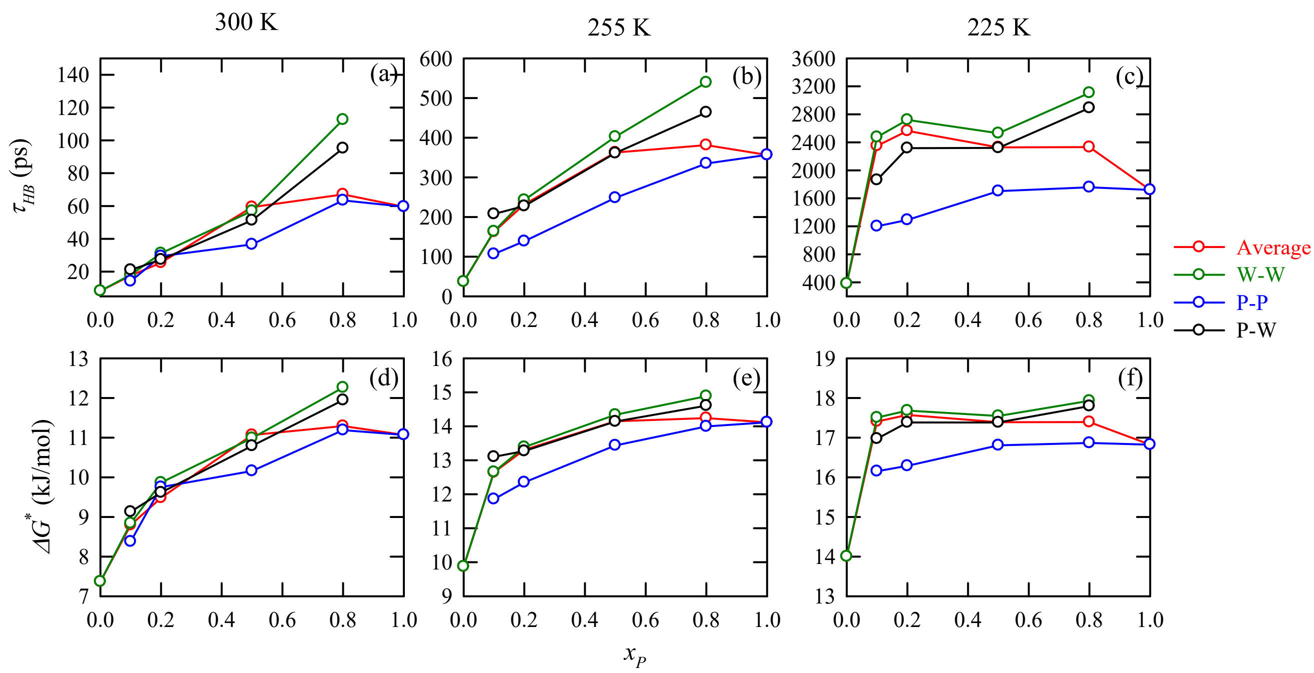

Figure S23. (a-c) H-bond lifetime, (d-f) free-energy of activation for breaking of H-bonds at three representative temperatures for the W/P binary mixture: 300 K (left panels), 255K (middle panels), and 225K (right panels).

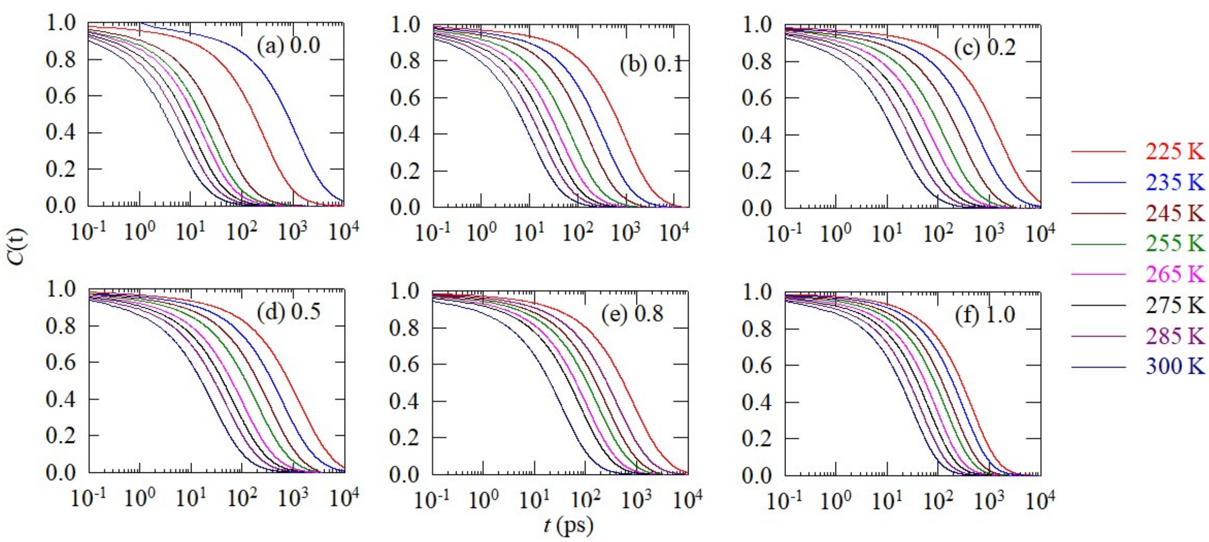

Figure S24. $C_{HB}(t)$ for the overall systems at all temperatures for W/E binary mixture.

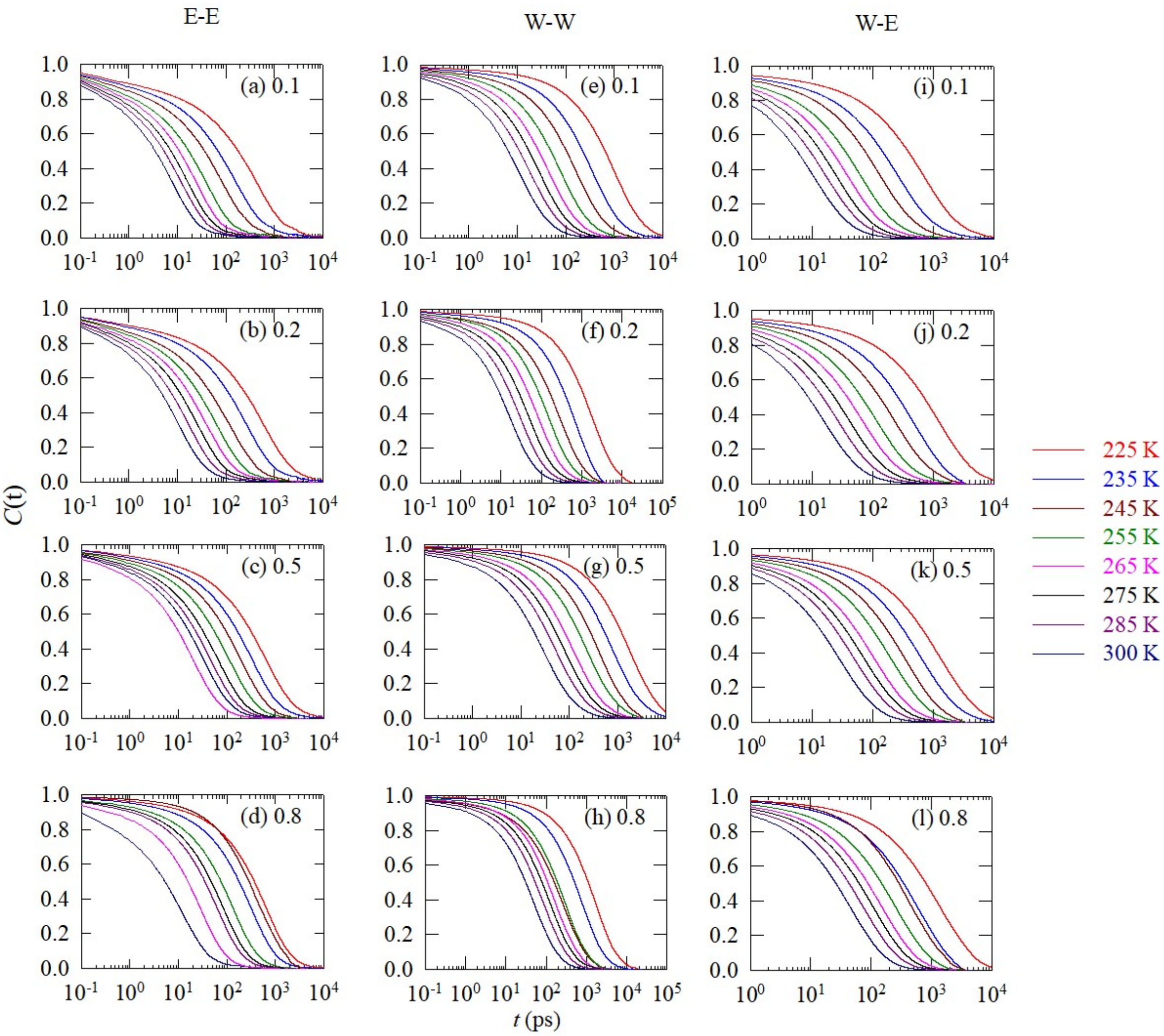

Figure S25. $C_{HB}(t)$ for the ethanol...ethanol, water…water, and water…ethanol systems at all temperatures for W/E binary mixture.

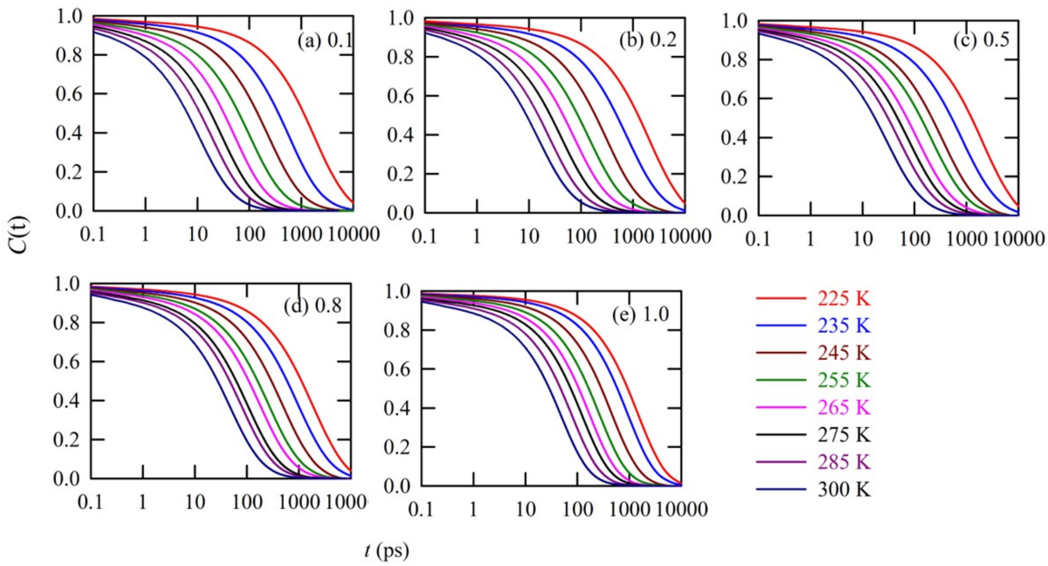

Figure S26. $C_{HB}(t)$ for the overall systems at all temperatures for W/P binary mixture.

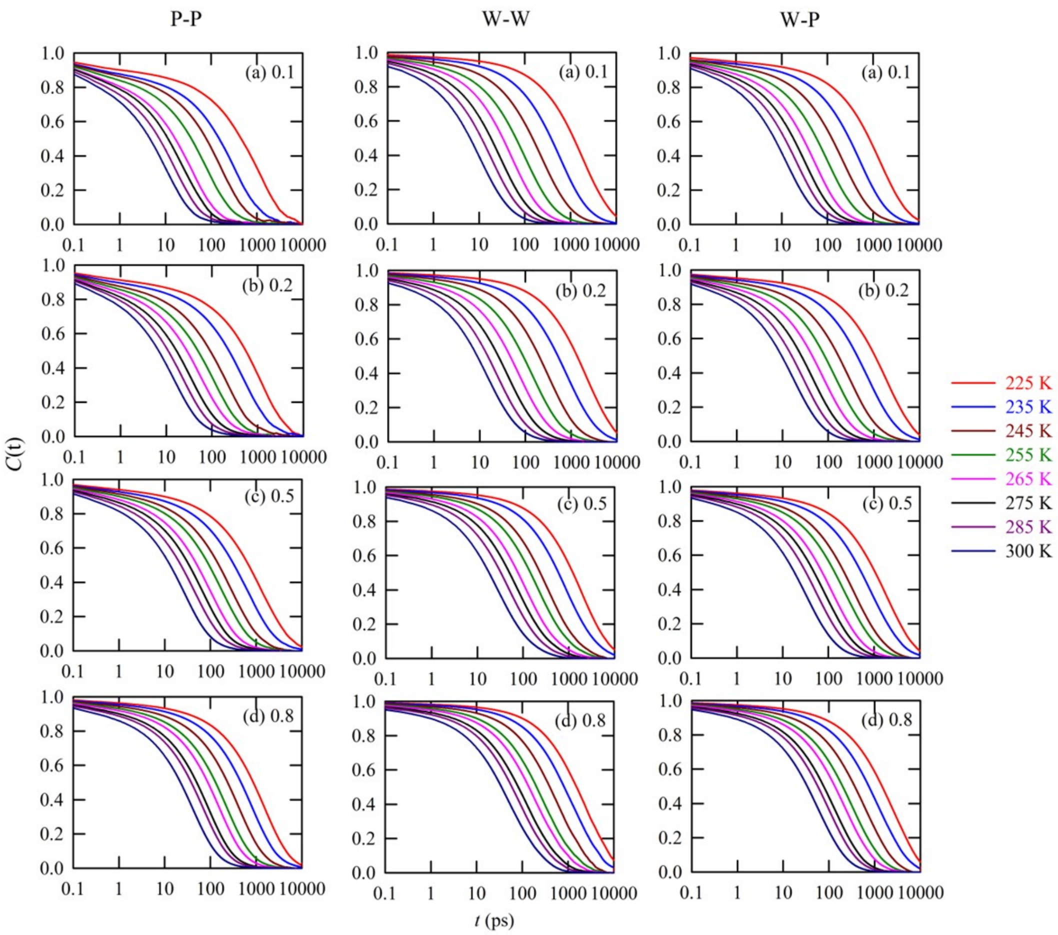

Figure S27. $C_{HB}(t)$ for the 1-propanol...1-propanol, water…water, and water…1-propanol systems at all temperatures.

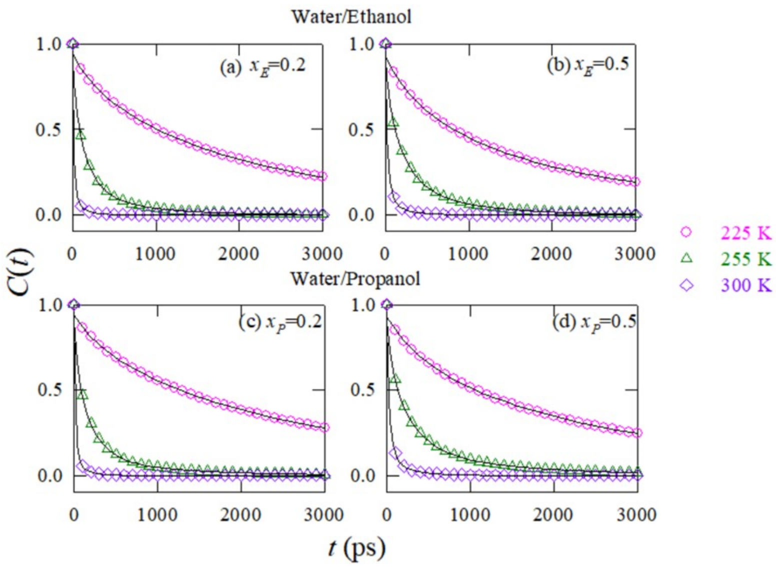

Figure S28. Fitting of $C_{HB}(t)$ for W/E and W/P mixtures for two representative alcohol mole fractions x=0.2 and 0.5 and three temperatures with the tri-exponential function,

$C_{HB}(t) = a_1 exp(-t/\tau_1) + a_2 exp(-t/\tau_2) + a_3 exp(-t/\tau_3); a_1 + a_2 + a_3 = 1$, shown by solid lines.

S2. Viscosity Analysis

The viscosity coefficient has been calculated using the Green– Kubo equation[2-5]

$$\eta = \frac{V}{10k_BT}\int_0^\infty \sum_{\alpha\beta}^{9} P_{\alpha\beta}(0)P_{\alpha\beta}(t)dt \tag{1}$$

In the above equation, k_B is the Boltzmann constant, V is the volume of the box, and T is the temperature of the system. The brackets ‘‘< >’’ refer to the ensemble average. $P_{\alpha\beta}$ is the pressure tensor, which can be calculated from the stress tensor $\sigma_{\alpha\beta}$ using the following equation,

$$P_{\alpha\beta} = \frac{1}{2}(\sigma_{\alpha\beta} + \sigma_{\beta\alpha}) - \frac{1}{3}\delta_{\alpha\beta}\left(\sum_{\alpha}\sigma_{\alpha\alpha}\right) \tag{2}$$

$\delta_{\alpha\beta}$ is the Kronecker delta. Note that in the above nine $P_{\alpha\beta}$ elements only six $P_{\alpha\beta}$ elements are distinct. Note that equation (1) improves the statistics over the original Green–Kubo relation, which considers the average of only three off-diagonal terms.

$$\eta = \frac{V}{3k_BT}\int_0^\infty \left\langle \sum_{\alpha\beta}^{3} P_{\alpha\beta}(0)P_{\alpha\beta}(t) \right\rangle dt \tag{3}$$

However, equations (1) and (3) do not result in significantly different η values when sufficiently long simulation trajectories are examined[6-8]. Figures S29 and S31 display the pressure auto-correlation functions at three different temperatures for the W/E and W/P mixtures at two alcohol mole fractions. Figures S30 and S32 show the limit of the integration in Eq. 3[9].

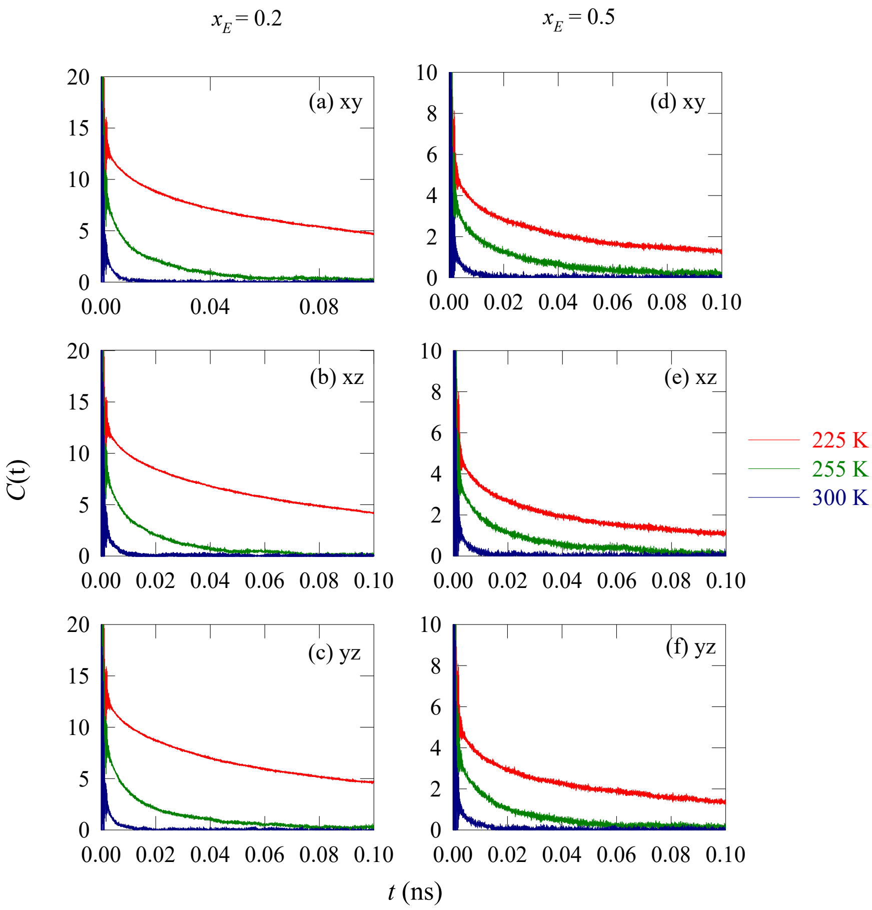

Figure S29. The pressure auto-correlation functions at three representative temperatures (225K, 255K, and 300K) for the W/E binary mixture with x_E=0.2 and 0.5 mole fractions of ethanol.

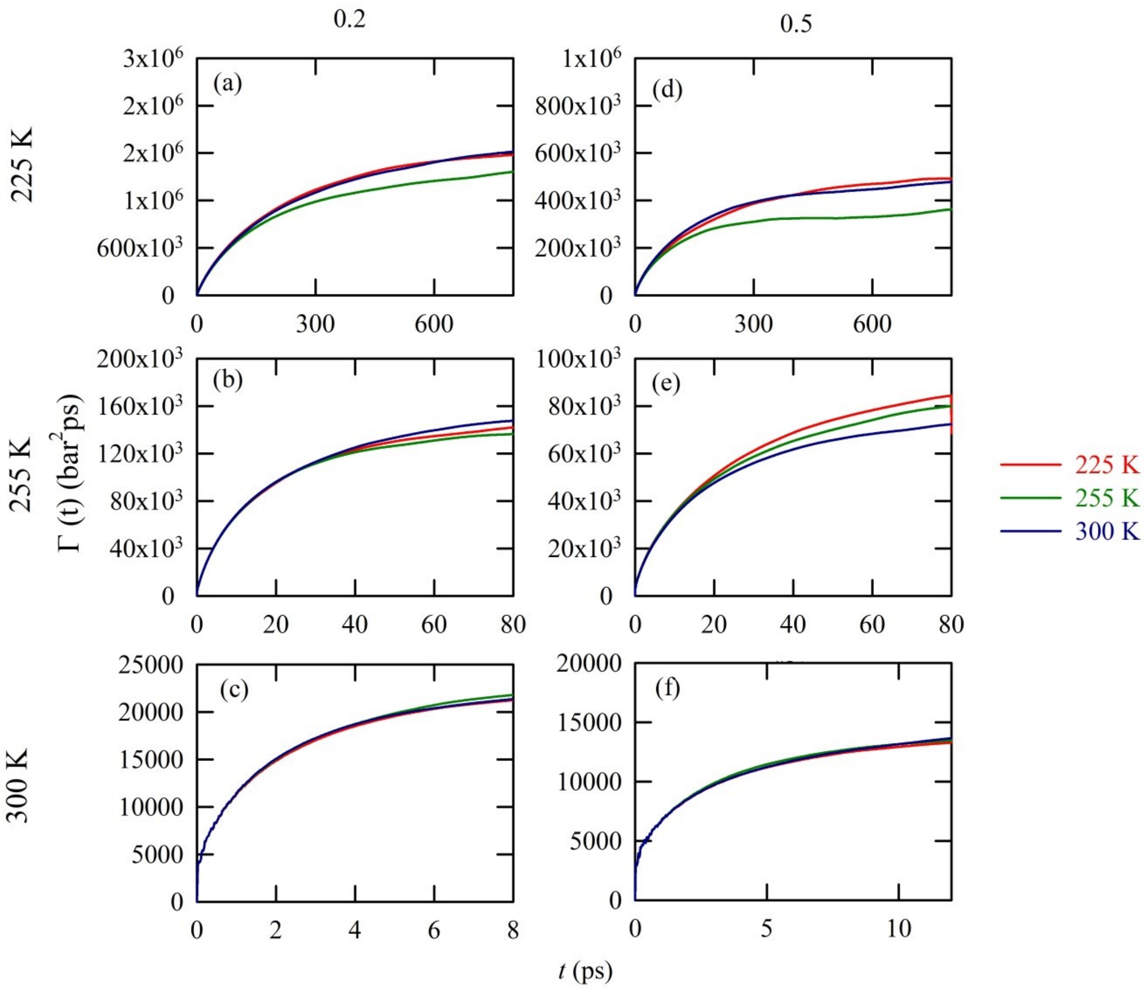

Figure S30. The integrated of correlation of viscosity at three representative temperatures (225K, 255K, and 300K) for the W/E binary mixture with x_E=0.2 and 0.5 mole fractions of ethanol.

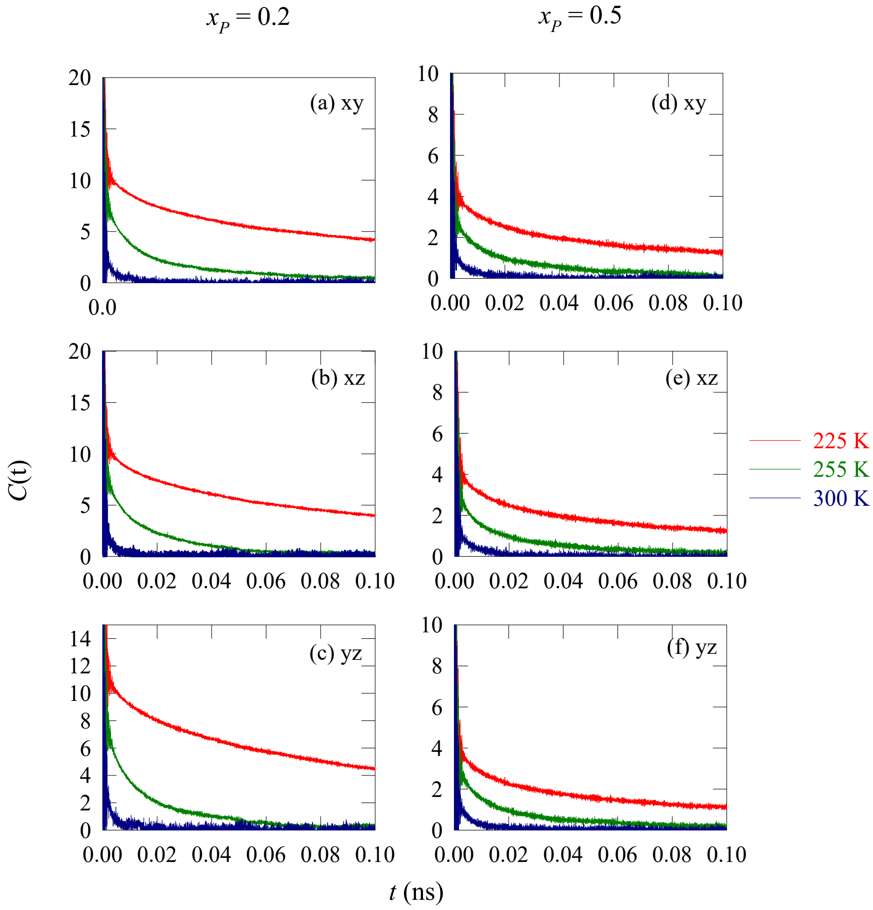

Figure S31. The pressure auto-correlation functions at three representative temperatures (225K, 255K, and 300K) for the W/P binary mixture with x_P=0.2 and 0.5 mole fractions of 1-propanol.

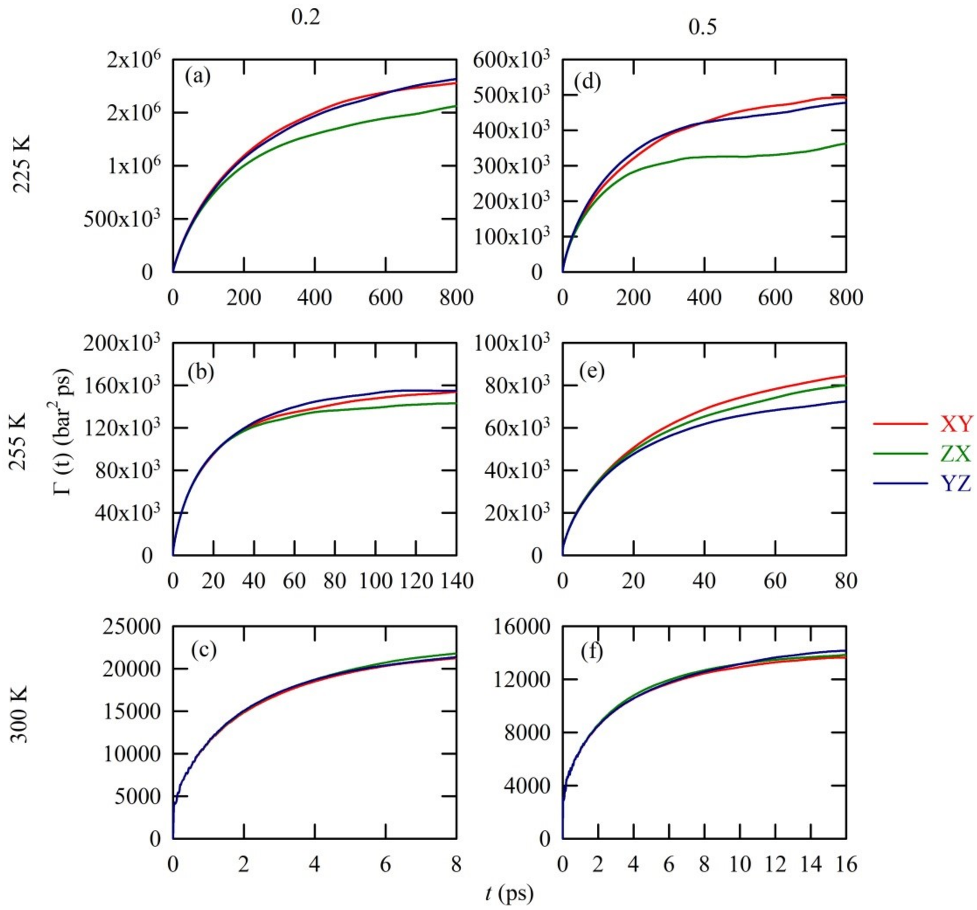

Figure S32. The integrated of correlation of viscosity at three representative temperatures (225K, 255K, and 300K) for the W/P binary mixture with *x*=0.2 and 0.5 mole fractions of 1-propanol.

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